UNIVERSITY OF ALBERTA

# LOW TEMPERATURE PROPERTIES OF THE ANTIFERROMAGNETIC FACE - CENTERED CUBIC ISING MODEL

by

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The undersigned certify that they have read, and recommend to the Faculty of Graduate Studies for acceptance, a thesis entitled LOW TEMPERATURE PROPERTIES OF THE ANTIFERROMAGNETIC FACE - CENTERED CUBIC ISING MODEL, submitted by Charles J. Elliott, in partial fulfilment of the requirements for the degree of Master of Science.

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## ABSTRACT

Seven terms of the low temperature partition function for the face-centered cubic antiferromagnetic lattice with Ising interactions have been calculated by counting bi-coloured graphical configurations. Low temperature series expansions for the specific heat, internal energy, and the zero field magnetic susceptibility are derived. The transition temperature $kT_c/J$ is estimated to be about $1.9 \pm .1$, obtained by extrapolating various series by Padé approximant and ratio methods. The specific heat curve was fitted to a logarithmic singularity at T and from the resulting curve an estimate of the critical entropy was obtained by direct integration of the area under the curve. We obtain $S_c = .312$ k.

Introduction

## 1.1 Cooperative Phenomena in Crystals

Macroscopic assemblies can be divided into two categories. In the first category, the microscopic subsystems of the assembly can be regarded as independent and non - interacting. Here the thermodynamic properties of the total system are determined by the energy levels of the microscopic subsystems and usually exhibit smooth and continuous behaviour. The second category includes systems in which the microscopic subsystems interact strongly. Here we find the phenomenon of phase transitions occuring, such as gas condensation, order-disorder transitions in alloys and ferromagnetism.

In 1925 Ising[1] proposed a simple model of such a system in which

(1) All atoms were identical but to each one there corresponds only two possible states.

(2) Only nearest neighbour (n.n.) interactions are significant.

He associated with each lattice point a spin coordinate $\sigma$ which could take on one of two possible values,

+1 for spin up and -1 for spin down. The interaction energy between the j and kth lattice points was postulated to be

$$E_{jk} = \begin{cases} -J\sigma_j\sigma_k & \text{if } j \text{ and } k \text{ are n.n.(nearest neighbour)} \\ 0 & \text{otherwise} \end{cases}$$

Thus the energy is -J if the n.n. atoms have parallel spins and +J if they have antiparallel spins.

In addition, if there is an external magnetic field, H, present each spin is coupled to the field by and interaction energy -m H if the spin is aligned with the field and +m H if the spin opposes the field.

m is the magnetic moment per spin.

We have then for the total energy in a lattice of N sites

$$E = -J \sum_{\substack{i,j=1 \\ j<i}}^{N} \sigma_i\sigma_j - H m \sum_{i=1}^{N} \sigma_i \tag{1}$$

and the partition function is

$$Z_N = \sum_{\sigma_1=\pm 1} \sum_{\sigma_2=\pm 1} \cdots \sum_{\sigma_N=\pm 1} \exp\left\{ \frac{J}{kT} \sum_{i,j=1}^{N} \sigma_i\sigma_j + \frac{mH}{kT} \sum_{i} \sigma_i \right\} \tag{2}$$

from which in principle all the thermodynamic properties can be derived.

## 1.2 Antiferromagnetism and the Face-centered Cubic Lattice

When J is negative we say that the system is antiferromagnetic.

In zero magnetic field the lowest energy states for a ferromagnet will be the two states in which all the spins are aligned. However, in the case of an antiferromagnetic lattice the lowest energy states will be the two in which the n.n. spins tend to be antiparallel.

For some lattices such as the simple quadratic (sq), simple cubic (sc) and body centered cubic (bcc) perfect antiferromagnetic ordering is possible (all spin up atoms are n.n. of spin down atoms only and vice versa). Clearly these "loose packed" lattices can be subdivided into 2 sublattices; one containing only "spin up" atoms and one only "spin down" atoms and nearest neighbour bonds exist between atoms in different sublattices only.

For other lattices including the triangular and f.c.c. such A.F. ordering is not possible since there exist closed paths in the lattice with an odd number of nearest neighbour bonds. Very little information is known about the thermodynamic properties of these lattices, especially in the low temperature region.

This thesis is concerned with the properties of the spin 1/2 Ising model of the f.c.c.. antiferromagnet.

Among the various lattices that have been studied extensively, the f.c.c. antiferromagnet seems to be unique in that it is the only close packed lattice that has been shown to have an order-disorder transition above 0°K. This has been recently shown by Betts and Elliott[2]; see also Section 3 in this thesis. Therefore it would be very interesting indeed, to know the exact nature of the transition point and any singularities which occur there and to compare these with other three dimensional loose packed antiferromagnet lattices. So far any available information has come from high temperature series expansions, but all these attempts do not produce any conclusive facts about low temperature behaviour. For example Burley[3], using the Kikuchi approximation, extrapolated series expansions for various lattices including the f.c.c. into the low temperature region. Sykes and Fisher[4], using improved techniques, have extrapolated exact series expansions of the loose packed sq and HC.(honeycomb) anti-ferromagnet lattices. Baker[5,6] found the Padé approximant method very successful in extrapolating antiferromagnet susceptibility series expansions of almost all the loose packed and close packed lattices. In 1961 Danielian [7] showed that two dimensional order existed in the f.c.c. antiferromagnet lattice and he calculated the ground state entropy. Later in 1964[8] he calculated the first three terms of the low temperature partition function expansion and gave a rough estimate of the transition temperature.

## 1.3 Scope of Thesis

Section 2 outlines the development of the exact series expansions which have been applied in subsequent sections. C. Domb[9] and Newell and Montroll[10] have contributed excellent review articles which cover most of the material in this section.

In section 3, the low temperature series expansions of the partition function and a long-range order parameter are derived for the f.c.c. antiferromagnet. A discussion of the configurational problems involved is also included.

In section 4, the critical temperature is estimated by extrapolating series expansions of the specific heat curve and the long range order parameter. The critical entropy is also estimated. Section 5 is a brief summary of the results obtained and conclusions.

# Theory

## 2.1 Low Temperature Series Expansions

The partition function can be expressed

$$Z_N = \sum_{N_1, N_{12}} g(N, N_1, N_{12}) \exp(-E(N, N_1, N_{12})/kT) \qquad (3)$$

where the conbinatorial factor $g(N,N_1,N_{12})$ specifies the number of possible configurations of the lattice corresponding to $N_1$ atoms with spin up and $N_{12}$ nearest neighbour bonds between unlike spins. E is the total energy of the microstate and depends only on $N, N_1$ and $N_{12}$.

An equivalent low temperature expansion of (3) can be formed by starting with the lattice in the ground state and enumerating all possible configurations as more and more spins have overturned.

Thus we can write

$$Z_N = \sum_{a,b} g(N,a,b) \exp(-E(N,a,b)/kT) \qquad (4)$$

where a = number of spins overturned

bJ = increase in energy resulting from the "a" overturned spins.

g(N,o,o) is then the degeneracy of the ground state configuration.

We first consider the ferromagnetic case ($J>o$). The total configurational energy E can be written

$$E = E_o + E_b$$

where $E_o$, the ground state energy is

$$E_o = -N\ m\ H - q\ \frac{N}{2}\ J$$

q = the number of nearest neighbours per atom.

$$E_b = bJ + 2a\ m\ H$$

Substituting in (4) we get

$$Z_N = z^{-q(N/4)}\ \mu^{-(N/2)} \sum_a \sum_b g(N,a,b)\ z^{b/2}\ \mu^{a/2} \qquad (5)$$

We have introduced the low temperature variables

$$z = \exp(-2J/kT)$$

$$\mu = \exp-(2mH/kT)$$

which tend to zero as $T \to 0$.

If we write the series in (5) as $\Lambda_N(\mu,z)$ then (5) is shortened to

$$Z_N = z^{-q(N/4)}\ \mu^{-(N/2)}\ \Lambda_N(\mu,z) \qquad (6)$$

In zero field $\mu = 1$ and

$$Z_N = z^{-q(N/4)}\ \Lambda_N(1,z)$$

For the loose packed antiferromagnetic lattices in zero field, there is an exact correspondence in energy between the configurations of a ferromagnet and those of the antiferromagnet. This means that the partition function in the absence of a magnetic field must be invariant under the transformation $J \to -J$ (see for example (9) page 175).

However, we will primarily be concerned with the close packed f.c.c. antiferromagnetic lattice which does not fall into this category. From here on all statements will refer to the f.c.c. Ising antiferromagnet unless otherwise indicated. Its partition function is not invariant under such a transformation.

Let us suppose that in the ground state in zero field every +1 spin is along the z direction (direction 1) and every -1 spin is antiparallel to the z direction (direction 2). We will further suppose that every +1 spin will have $q_{11}$ nearest neighbours which are also +1 spin and $q_{12}$ nearest neighbours which are spin -1 (the subscripts referring to the direction of each spin).

The following relations are then true

$$q_{11} + q_{12} = q_{22} + q_{21} = q$$

$$q_{12} = q_{21}$$

$$q_{22} = q_{11}$$

It also follows that there will be N/2 +1 spins and N/2 -1 spins. The above conditions are not necessarily true for all close packed lattices. The two dimensional triangular lattice is an example in which they are not[11].

In a strong magnetic field the ground state must become ferromagnetically ordered but for weak fields the ground state will be unchanged.

The critical field $H_c$ occurs when these two states have equal energy i.e.

$$\frac{qN}{2}(-J) - mN\,H_c = \frac{N}{2}(-Jq_{11} + q_{12}\,J) \tag{8}$$

From equation (8) we can determine the critical field

$$mH_c = -q_{12}\,J \qquad (J<o) \tag{9}$$

We will now consider the low temperature partition function expansion for fields less than $H_c$.

We have

$$E_o = \frac{NJ}{2}(q_{12} - q_{11})$$

$$= NJq'$$

$$\text{where} \quad q' = q_{12} - \frac{q}{2}$$

$$E_b = -bJ - 2m\,H\,(a_1 - a_2)$$

where

$a_1$ = no. of 1 spins overturned

$a_2$ = no. of 2 spins overturned

$$\therefore Z_N = \sum_{a,a_1} \sum_b g(N,a,b) \exp[(-Nq' J + bJ + 2m H(a_1-a_2))/kT]$$

$$Z_N = y^{-Nq'/2} \sum_{a,a_1} \sum_b g(N,a,b)\, y^{b/2}\, \mu^{-(a_1-a_2)} \qquad (10)$$

where y = exp(2J/kT) is the antiferromagnetic low temperature variable which tends to zero as $T \to 0$. We will make use of (10) later. As an illustration, the term corresponding to two overturned spins; one in direction 1 and the other in direction 2 is

$$y^{-Nq'/2} [g(N,2,b)(\mu^{-1} + \mu^{+1})\, y^{b/2}] \qquad (11)$$

## 2.2 High Temperature Series Expansions*

Starting from equation (2) in the form

$$Z_N = \sum_{\sigma_i=\pm 1} \prod_{nn} \exp(k\, \sigma_i\sigma_j) \prod_i \exp(I\, \sigma_i)$$

it can be shown that this reduces to

$$Z_N = (\cosh K)^A (\mathrm{Cosh}\ I)^N\, 2^N \sum_{\ell=0}^{qN} \sum_{m=0}^{2\ell} G(\ell,m,N) v^m u^\ell \qquad (12)$$

* see references (9) and (10)

where $A$ = number of nearest neighbour bonds

$K = J/kT$

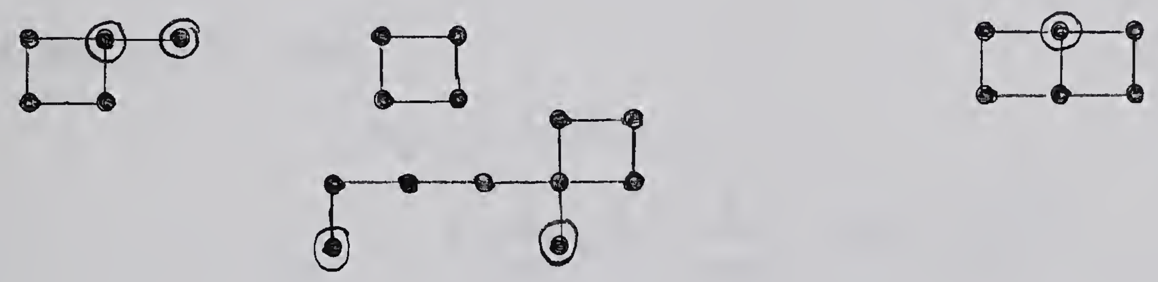
$u = \tanh K$

$I = mH/kT$

$v = \tanh I$

$G(\ell,m,N)$ is the combinatorial factor corresponding to the number of graphs of $\ell$ bonds and $m$ distinct spins; the $m$ spins coinciding with the odd vertices. An odd vertex is a vertex at which an odd number of bonds meet.

FIGURE I



$m = 2$, $\ell = 5$ (a) $m = 2$, $\ell = 13$ (b) $m = 1$, $\ell = 6$ (c)

In Figure 1, in which the spins are circled, (a) and (b) contribute but (c) does not.

The expansion (12) is a high temperature series expansion, since as $T \to \infty$ $u$ and $v \to 0$. (12) is valid for an antiferromagnet. Under the transformation $J \to -J$

$$[u(J)]^{\ell} \to (-1)^{\ell} [u(+J)]^{\ell}$$

In zero field the partition function reduces to a series involving closed polygons only.

## 2.3 Thermodynamic Properties

The thermodynamic properties are obtained directly from the series expansions of the partition function. Some of the thermodynamic variables are:

The internal energy per spin

$$U = kT^2 \frac{\partial}{\partial T} \frac{\ln Z_N}{N} \tag{13}$$

The specific heat

$$C_v = (\partial U/\partial T)_v$$

$$= \frac{\partial}{\partial T}(kT^2 \frac{\partial}{\partial T}) \quad (\ln Z_N)/N \tag{14}$$

The entropy per spin

$$S = \int_0^T dT \frac{C_v}{T} \tag{15}$$

The magnetic properties can also be obtained. The magnetization per spin along the z direction is the average induced magnetic moment per spin and is given by

$$M = kT \frac{\partial}{\partial H} \frac{\ln Z_N}{N} \tag{16}$$

The susceptibility per spin is

$$\chi = \partial M/\partial H \tag{17}$$

It is more convenient to express these variables in terms of y and $\mu$. Using the relation

$$T \frac{\partial}{\partial T} = -(\log y)y \frac{\partial}{\partial y} \tag{18}$$

the thermodynamic variables become

$$U = -\ 2J\ y \frac{\partial}{\partial y}\ (\ln Z_N)/N \tag{19}$$

$$\frac{C_V}{k} = (\log y)^2\ y\ [\partial/\partial y + y(\partial^2/\partial y^2)](\log Z_N/N) \tag{20}$$

$$S = -\int_0^y [Cv/(y\ \ln\ y)]\ dy \tag{21}$$

$$M = -2m\ \mu(\partial/\partial\mu)\ (\log Z_N/N) \tag{22}$$

$$\chi = \frac{(2m)^2}{kT}\ \mu[\partial/\partial\mu + \mu(\partial^2/\partial\mu^2)](\log Z_N/N) \tag{23}$$

The zero field susceptibility.

$$\chi_o = \lim_{H\to 0} \chi \tag{24}$$

# 3. Applications to the F.C.C. Antiferromagnet

## 3.1 Introduction

An exact low temperature series expansion for the partition function and a low temperature expansion for a long range order parameter has been calculated by configuration counting methods.

Danielian [7] has completely classified the ground state, showing that it was highly degenerate but the ground state entropy per spin was zero. He showed that two dimensional ordering in one of three perpendicular planes must exist. Recently it was shown [2] that the two dimensional order can be expressed by a three dimensional long range order parameter averaged over the statistical ensemble. The transition temperature has been estimated by finding the temperature at which the order disappears.

## 3.2 The Ground State

According to Danielian [7] the nature of the ground state can be elucidated as follows:

The f.c.c. lattice of N sites can be subdivided into N tetrahedra such that each bond is contained in one and only one tetrahedron.

Thus there are a total of 6N bonds. In the ground state each tetrahedron must be in its own ground state; i.e. two spins must be up and two down. Thus the configurational ground state energy is +2NJ. The degeneracy of the ground state is seen most easily by considering the lattice as being built up of layers of two dimensional quadratic lattices so that each atom of one layer is located above the middle of a square in the lower layer and each atom in the lower square is a nearest neighbour of the atom centered above that square. This is illustrated in Figure 2.

F.C.C. ANTIFERROMAGNETIC GROUND STATE

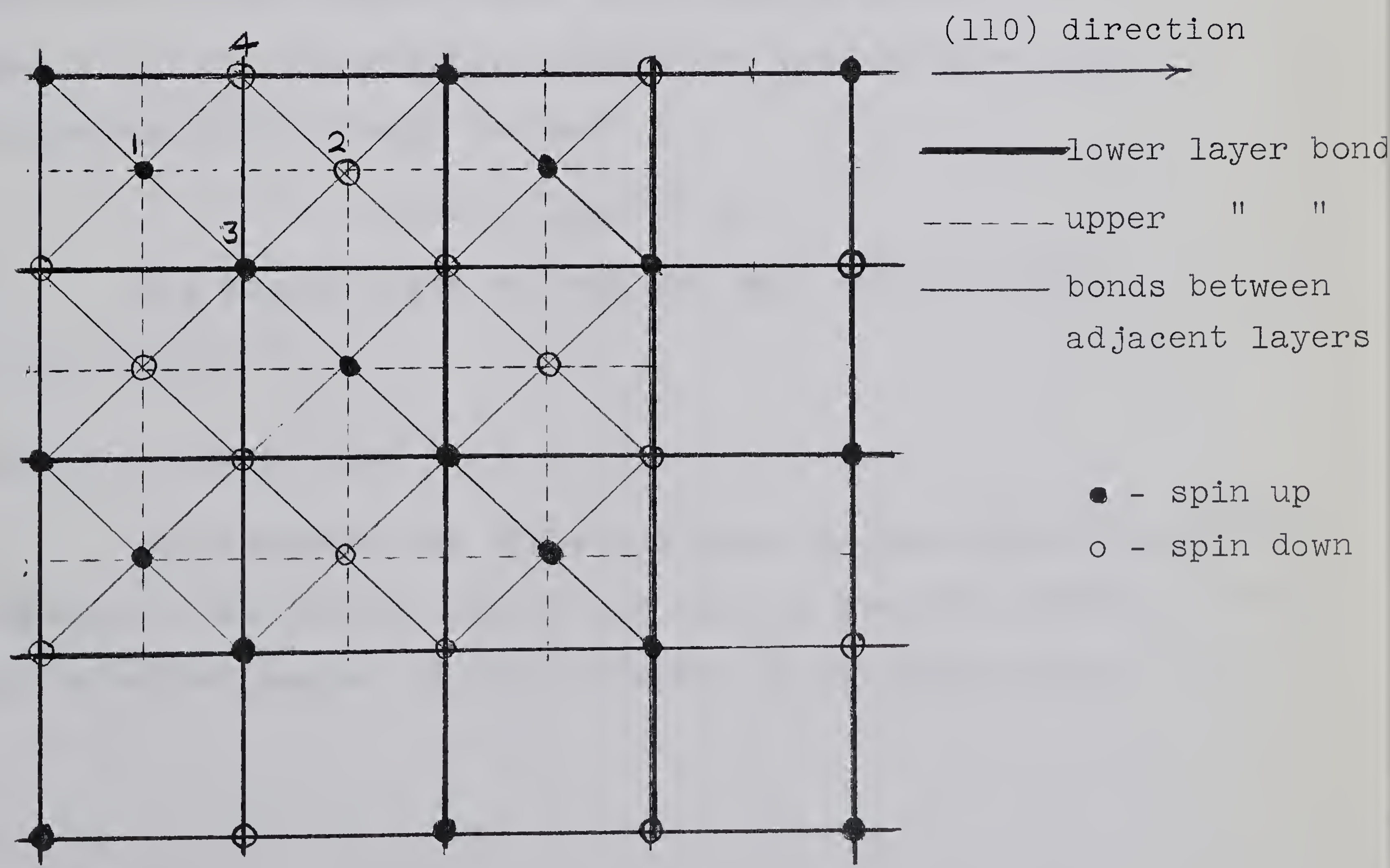



FIGURE 2

A partial layer of 25 atoms is shown and 9 atoms of the next layer. If we begin construction of the lattice with the lower layer antiferromagnetically ordered (alternating black and white) then when we arbitrarily add an atom to the next layer its spin state completely determines the states of all the atoms of that layer. Referring to Figure 2, if we choose atom 1 of the upper layer black then atom 2 must be white since atoms 1,2,3 and 4 form a tetrahedron in which we can only have 2 black atoms.

We conclude that the ground state consists of layers of antiferromagnetically ordered quadratic nets, also each atom has 4 like nearest neighbours and 8 unlike nearest neighbours. There are $(2N)^{1/3}$ such layers and so the degeneracy of the ground state is in the order of $2^{(2N)^{1/3}}$ since there are two possible states for each ordered plane. The ground state entropy is then

$$S(o) = k(2N)^{1/3} \ln 2$$

The ground state entropy per spin tends to zero in the limit $N \to \infty$.

## 3.3. Long Range Order

We designate two sites a,b along a line connecting nearest neighbour atoms in the lattice and specify that the distance between them be an odd number of bond lengths. If the expectation $< \sigma_a \sigma_b >$

is non zero we say that order exists and if $< \sigma_a \sigma_b >$ is non zero in the limit distance $a - b \to \infty$ then long range order is present.

For the f.c.c. antiferromagnet we know that in the ensemble of states at T = 0°K one third of all possible a and b sites will be in the ordered planes. For these cases $\sigma_a \sigma_b = -1$. The expectation $< \sigma_a \sigma_b >$ for all other cases we would expect to be zero from the way in which the ground state is constructed. The expectation $< \sigma_a \sigma_b >$ for any a, b then is not zero but exactly $-1/3$. We can conclude then, that "partial" three dimensional order exists.

The parameter $\sigma_a \sigma_b$ is not symmetrical in three dimensions since we do not know if a, and b lie in the ordered plane or in one of the other two. However, we can construct an order parameter that is symmetrical in all 3 directions. Consider a large regular tetrahedron in the lattice built up of smaller tetrahedra, and with its edges parallel to the nearest neighbour directions. If each edge contains an even number of spins, the following property is evident. In the ground state a pair of opposite edges of the tetrahedron will lie on parallel planes. Since there are three pairs of opposite edges, corresponding to the three sets of mutually perpendicular planes, one of the pairs must lie in the ordered plane. As a consequence

of the alternation of spins along these edges, two of vertices will have "spin up" and two "spin down".

Let $< t_{22} >$ represent the mean fraction of tetrahedra of a given size with two spins up and two spins down. Also define

$$< \alpha_1 > \ = \text{mean fraction of spins up}$$

$$< \alpha_2 > \ = \text{mean fraction of spins down.}$$

Clearly, then if there is no correlation among the spins of the 4 vertices of the tetrahedron we must have

$$<t_{22}> \; = \; 6 < \alpha_1 >^2 < \alpha_2 >^2 \tag{24}$$

since there are 6 distinct ways of labelling the 4 spins. Following the method of Domb[9] we modify (24) to take care of the correlation by introducing an order parameter $R_4$ as follows

$$< t_{22} > \; = \; 6 < \alpha_1 >^2 < \alpha_2 >^2 \left(1 + \frac{5}{3} R_4\right) \tag{25}$$

If $R_4 = 0$ there is no correlation. For a zero field configuration $< \alpha_1 > \; = \; < \alpha_2 > \; = 1/2$ so we can write

$$5/3\ (R_4) = 8/3\ (< t_{22} >) - 1 \tag{26}$$

and we see that long range order disappears when $< t_{22} >$ decreases to 3/8.

In the following equations $R_4$ is expressed in terms of the Ising variables $\sigma_a, \sigma_b, \sigma_c$, and $\sigma_d$ where a,b,c and d denote the 4 vertices of the tetrahedron.

$$t_{22} = \frac{3}{8} - \frac{1}{8}(\sigma_a\sigma_b + \sigma_b\sigma_c + \sigma_c\sigma_d + \sigma_a\sigma_c + \sigma_a\sigma_d + \sigma_b\sigma_d) + \frac{3}{8}(\sigma_a\sigma_b\sigma_c\sigma_d) \tag{27 a}$$

Also

$$R_4 = -\frac{1}{5}(\sigma_a\sigma_b + \sigma_b\sigma_c + \sigma_c\sigma_d + \sigma_a\sigma_c + \sigma_a\sigma_d + \sigma_b\sigma_d) + \frac{3}{5}(\sigma_a\sigma_b\sigma_c\sigma_d) \tag{27 b}$$

The right side of (27 a) is always unity for two plus and two minus spins and zero otherwise. Equation (26) will be used later to determine the Curie point defined as the temperature at which $R_4 \to 0$.

It was mentioned earlier that $<\sigma_a\sigma_b>$ is equal to $-1/3$ in the ground state and equal to zero at the transition temperature. This suggests a natural definition of a two spin order parameter.

$$R_2 = -3 < \sigma_1\sigma_2 > \tag{28}$$

Although $R_2$ lacks the symmetric features of $R_4$ it can still be thought of as a three dimensional order parameter as long as we do not specify the planes in which

sites 1 and 2 lie. In fact it is possible to show that $R_2$ and $R_4$ are related by the equation

$$5R_4 = 2R_2 + 3(R_2)^2 \tag{29}$$

Equation (29) is derived from a relation between the Ising variables $< \sigma_1\sigma_2 >$ for 2 spins and $< \sigma_1\sigma_2\sigma_3\sigma_4 >$ for 4 spins. We have

$< \sigma_1\sigma_2\sigma_3\sigma_4 > = 1/9 \ < \sigma_1\sigma_2 >^2$ since two of the four spins of the tetrahedron are independent of the spin states of the other two.

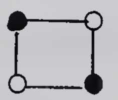
We can see from (29) that in the range $0 \leq R_2 \leq 1$, we have $R_2 \geq R_4$, the equality being true only at the end points. Physically this means that the correlation between two spins is always greater than the correlation between four spins which is what one would expect.

For the calculation of $T_c$, the parameter $R_4$ was used although $R_2$ would lead to the same result.

## 3.4 Low Temperature Series Expansions

The first few terms of the expansion series equation (10) and their corresponding graphs and contributions are listed in Table I, below:

TABLE I

| TERM | GRAPH | CONTRIBUTION |
|---|---|---|
| g(N,o,o) | | 1 |
| g(N,1,8) | ●, ○ | $\frac{N}{2}(\mu + \mu^{-1})$ |
| g(N,2,12) | ●—○ | 4 N |
| g(N,2,16) | ○ ○, ● ● | $(1/4)(N^2 - 10N)(\mu^2 + \mu^{-2})$ |
| | ● ○ | $(1/4)(N^2 - 16N)$ |
| g(N,3,16) | ●—○—●, ○—●—○ | $10N(\mu + \mu^{-1})$ |
| g(N,4,16) |  | $(3/2)N$ |

LOWEST ENERGY OVERTURNED SPIN CONFIGURATIONS

The configuration increase in energy -bJ is given by

$$\Delta E = -8nJ - 4J\,\delta_n{}^* \qquad (30)$$

where n = number of spins in the configuration

$\delta_n$ = number of even bonds less the number of odd bonds (an even bond connects like spins and an odd bond connects unlike spins).

The smallest change in energy is obtained by overturning one spin and is equal to -8J. This gives rise to the factor $(N/2)\mu\, y^4$ for an overturned black spin and $(N/2)\mu^{-1}y^4$ for an overturned white spin. The next smallest change of energy possible is -6J, produced by overturning adjacent black and white spins. The number of configurations increases rapidly for high powers of y. All the antiferromagnet low temperature configurations for the f.c.c. up to 5 spins and all others corresponding to an energy change of $\Delta E$ = 14J or less have been enumerated in Appendix A. Table II on the next page shows how the number of topologically different graphs increases with $\Delta E$.

* (30) follows directly from the ground state conditions that every spin has 4 like neighbours and 8 unlike neighbours.

TABLE II

| $\Delta E/2$ | Number of Distinct Graphs |
|---|---|
| 0 J | 0 |
| 4 J | 1 |
| 6 J | 1 |
| 8 J | 3 |
| 10 J | 8 |
| 12 J | 31 |
| 14 J | 127 |

NUMBER OF OVERTURNED SPIN CONFIGURATIONS

## 3.5 Configuration Counting

The number of ways a graph can be placed on a lattice of N sites divided by N is called the "lattice constant" for that graph. The enumeration of all the graphs and the calculation of the lattice constants is a difficult and time consuming task. Domb, in his review article[9] has outlined some of the standard techniques for counting, therefore, only the difficulties encountered with antiferromagnetic f.c.c. configurations will be pointed out here.

The first problem in obtaining the low temperature series expansion is to enumerate all the graphs corresponding to each increment of energy. This is easy enough for low energy graphs but is liable to error for higher energy graphs as a glance at Table II. will indicate. One approach is to assume that all the graphs have been accounted for when one can not think of any more. However, equation (30) seems to offer a more systematic approach.

For example, suppose we want to enumerate all possible 6 spin configurations that produce an energy change of $\Delta E = 28J$ (excluding any magnetic contribution) when the spins are overturned. Equation (30) states

$$\delta_6 = + 7 - 2 = 5$$

i.e. The number of odd bonds must be five greater than the number of even bonds. So the only possibilities are (5,0), (6,1), (7,2), .... etc. where the numbers in brackets are the numbers of odd and even bonds respectively. We begin with (5,0) and we notice that the only possible colourings for the f.c.c. are 2b, 4w; 2w, 4b; and 3b, 3w. The first two colourings are equivalent. We consider each colouring separately and group all of our graphs accordingly.

In the group G[(5,0) 3b, 3w] we find the following set of graphs:

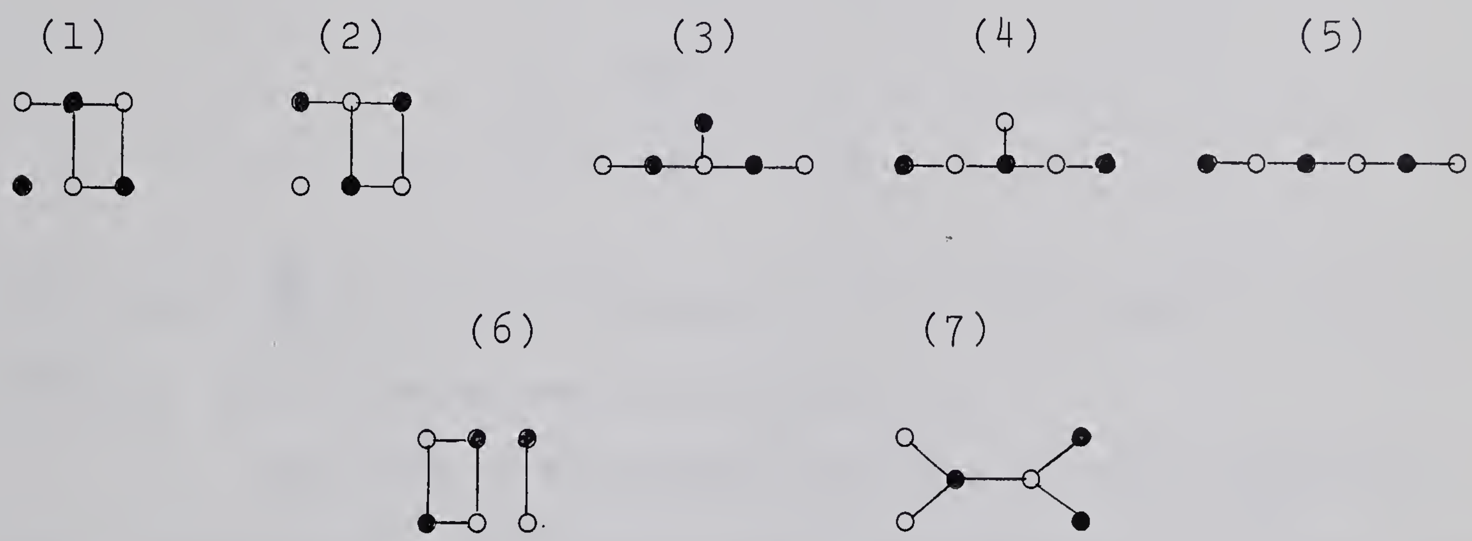



Figure 3

and we wish to know if it is complete.

If the 6 vertices are all considered to be distinguishable (numbered for instance), then the number of ways of adding the 5 odd bonds is clearly ${}_9C_5$ or 126.

Now define the order P of each graph as

$$P = \frac{n_b!\ n_w!}{S}$$

where $n_b$ = number of black vertices

$n_w$ = number of white vertices

S is a symmetry number which is equal to the number of automorphisms of the labeled graph.

In our example

$$P_1 = P_2 = \frac{(3!)^2}{2} = 18$$

$$P_3 = P_4 = \frac{(3!)^2}{2} = 18$$

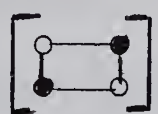
$$P_5 = \frac{(3!)^2}{1} = 36$$

$$P_6 = \frac{(3!)^2}{4} = 9$$

$$P_7 = \frac{(3!)^2}{4} = 9$$

The total $\sum_{i=1}^{7} P_i = 126$ showing that all the graphs in group G[(5,0) 3b 3w] have been accounted for.

The only discouraging feature of this method is that some configurations that are incompatible with the lattice structure must also be included.

The task of determining the lattice constants for each configuration is complicated by the degeneracy of the ground state. For example, the number of times the configuration  appears in the lattice is dependent upon which particular ground state of the ensemble of ground states that the lattice is in. Consequently it is necessary to use elementary probability considerations.

## 3.6 Long Range Order Series Expansion

Returning to equation (26), which defines the long range order parameter $R_4$, we can compute the expectation $< t_{22} >$ by taking an ensemble average as follows:

$$< t_{22} > = \frac{\sum_b g(N,b,t_{22})\ y^{b/2}}{\sum_b g(N,b)\ y^{b/2}} \tag{31}$$

where

$$g(N,b,t_{22}) = \begin{cases} 0 & \text{if } t_{22} = 0 \\ g(N,b) & \text{if } t_{22} = 1 \end{cases}$$

Thus $g(N,b,t_{22})$ counts the number of ways each configuration can be placed on the lattice so that the left side of (27) is one after overturning the spins. Clearly the only possibilities are

1) none of the 4 vertices are overturned
2) one w and one b vertex are overturned
3) two w and two b vertices are overturned

The four vertices are assumed to be infinitely far apart so that no closed configurations can overlap two vertices simultaneously.

## 3.7 Counting Theorem

A relationship between the terms in equation (31) to the terms in the partition function has been rigorously

established, and considerably simplifies computation.

We will adopt the following notation:

Let $X_i$ represent a connected configuration

$\{X_i\}$ - the contribution of X to the coefficient in the partition function

$[X_i]$ - the contribution of X to the coefficient in the expansion series for (8/5) $R_4$.

$X_i X_j X_k$ - a configuration with three components.

$$\{X_1 X_2 \text{ -- } X_i \text{ -- } X_M\} = \{X_i\}_M$$

$$\left[X_1 X_2 \text{ -- } X_i \text{ -- } X_M\right] = [X_i]_M$$

are similar contributions from configurations of M components.

$\overset{\circ}{X}_i$ - a graph which coincides with one of the two white vertices of the tetrahedron. We will refer to such graphs as being "rooted".

Thus

$[\overset{\circ}{X}_i \overset{\bullet}{X}_j \overset{\bullet}{X}_k]_M$ is the number of ways the disconnected graph of M components can be put on the lattice so that two black vertices and one white vertex of the tetrahedron are overturned. Note that it is impossible for one connected graph to be rooted to two vertices simultaneously since the vertices are assumed to be infinitely far apart.

The counting theorem then states for a disconnected configuration of M distinct components

$$[X_i]_M = \{X_i\}_M - (4/N)\, n_i \{X_i\}_M$$

$$+ \frac{8}{N^2} \{X_i\}_M \sum_{ij} \frac{\{X_i\}\ \{X_j\}}{\{X_i\ X_j\}} (W_iW_j+B_iB_j+2W_iB_j)\lambda_{ij}$$

$$- \frac{6}{N^3} \{X_i\}_M \sum_{i,j,k} \frac{\{X_i\}\{X_j\}\{X_k\}}{\{X_iX_jX_k\}} (W_iW_jB_k+B_iW_jW_k)\ \lambda_{ijk}$$

$$+ \frac{384}{N^4} \{X_i\}_M \sum_{i,j,k,} \frac{\{X_i\}\{X_j\}\{X_k\}\{X_\ell\}}{\{X_iX_jX_kX_\ell\}} (W_iW_jB_kB\ )\lambda_{ijk\ell} \qquad (32)$$

The corresponding counting theorem for the coefficients of the expansion series of $(2/3)R_2$ is

$$[X_i]_M = \{X_i\}_M - (2/N)\, n_i \{X_i\}_M$$

$$+ (4/N^2)\ \{X_i\}_M \sum_{i,j} \frac{\{X_i\}\{X_j\}}{\{X_iX_j\}} (W_iW_j+B_iB_j)\ \lambda_{ij} \qquad (33)$$

where $W_i$ = the number of white vertices of $X_i$

$B_j$ = the number of black vertices of $X_j$

$n_i$ = the number of vertices for $X_i$

The summations in (32) and (33) are taken over all different combinations of the M components so that in each term $i \neq j \neq k \neq \ell$ . The $\lambda$'s are factors to take care of cases in which the M components are not all distinct. The proof of the counting theorem is given in Appendix B along with the values of the $\lambda$ factors.

# 4. Thermodynamic Properties

## 4.1 Low Temperature Series Expansion of Thermodynamic Variables

Applying the methods described in sections 2 and 3, the following low temperature expansion for the partition function in a magnetic field was obtained.

$$
\begin{aligned}
Z_N = 2^{(2N)^{1/3}}\, y^{-N} \{ & 1 + (N/2)\,(\mu+\mu^{-1})y^4 + 4Ny^6 + [(N^2/4) - 5/2\, N \\
& + 10N\,(\mu+\mu^{-1}) + ((N^2/8) - (5/4))(\mu^2+\mu^{-2})]y^8 \\
& +[(209/2)N + (2N^2-27N)(\mu+\mu^{-1}) + 9N(\mu^2+\mu^{-2})]y^{10} \\
& +[18N^2 - (619/2) + (1/16)(N^3 - 30N^2 + 6735N)(\mu+\mu^{-1}) \\
& +(5N^2 - 85N)(\mu^2+\mu^{-2}) + (1/48)(N^3 - 30N^2 + 320N)(\mu^3+\mu^{-3})]y^{12} \\
& +(1/8)[8N^3 - 296N^2 + 38265N + (774\, N^2 - 16049\, N)(\mu+\mu^{-1}) \\
& +(4N^3 - 148N^2 + 7290N)(\mu^2+\mu^{-2})+(36N^2 - 768N)(\mu^3+\mu^{-3})\,]y^{14} \\
& + \text{--------} \} \qquad (34)
\end{aligned}
$$

On physical grounds

$$\lim_{N\to\infty} Z_N \to [Z(y)]^N$$ where $Z(y)$ is the

partition function per spin.

$$\log Z = \lim_{N\to\infty} [(1/N) \log Z_N(y,N)] \tag{35}$$

We get

$$\log Z = -\log y + \{(1/2)(\mu+\mu^{-1})\, y^4 + 4y^6$$

$$+[-(5/2) + 10\,(\mu+\mu^{-1}) - (5/4)^{*}\,(\mu^2+\mu^{-2})]y^8$$

$$+[(209/2) - 27\,(\mu+\mu^{-1}) + 9(\mu^2+\mu^{-2})]y^{10}$$

$$+[-(619/2) + (6735/16)(\mu+\mu^{-1}) - 85(\mu^2+\mu^{-2})$$

$$+ (20/3)\,(\mu^3+\mu^{-3})]y^{12} + (1/8)[38265 - 16049\,(\mu+\mu^{-1})$$

$$+ 7290\,(\mu^2+\mu^{-2}) - 768\,(\mu^3+\mu^{-3})]y^{14} + \text{------}\} \tag{36}$$

* Equation 5 in reference 8 is in error for this coefficient.

In zero field the partition function reduces to

$$Z = -\log y + y^{4} + 4y^{6} + 15y^{8} + (137/2)\, y^{10}$$

$$+(9017/24)\, y^{12} + (19211/8)\, y^{14} + \text{-------} \qquad (37)$$

From (36) and (37) we derive the low temperature series expansions for the thermodynamic variables as described in Section 2.

Internal Energy per Spin

$$U = 2J\{\, 1 - 4y^{4} - 24y^{6} - 120y^{8} - 685\, y^{10} - (9017/2)\, y^{12}$$

$$- (134477/4)\, y^{14} \text{------}\} \qquad (38)$$

Zero Field Specific Heat

$$(C_v/k) = (\log y)^{2} \ \{16y^{4} + 144y^{6} + 960y^{8} + 6850y^{10} +$$

$$+ 54102y^{12} + (941339/2)\, y^{14} + \text{--------}\} \qquad (39)$$

Magnetization

$$M = -2m\, y^{-N} \ \{(N/2)(\mu-\mu^{-1})y^{4}$$

$$+[10N(\mu- \mu^{-1}) + ((N^{2}/4) - (5/2)N)(\mu^{2}-\mu^{-2})]y^{8}$$

$$+[(2N^2 - 27N)(\mu-\mu^{-1}) + 18N(\mu^2-\mu^{-2})]y^{10}$$

$$+[(1/16)(N^3 - 30N^2+6735N)(\mu-\mu^{-1})+(10N^2-170N)(\mu^2-\mu^{-2})$$

$$+(1/16)(N^3 - 30N^2 + 320N)(\mu^3-\mu^{-3})]y^{12}$$

$$+(1/8)[(774N^2-16049N)(\mu-\mu^{-1})+(8N^3-296N^2+14580N)$$

$$(\mu^2-\mu^{-2})+(108N^2 - 2304N)(\mu^3-\mu^{-3})]y^{14}+ ------\} \qquad (40)$$

Magnetization per Spin

$$M = -2m \{(1/2)(\mu-\mu^{-1})y^4$$

$$+[10(\mu-\mu^{-1}) - (5/2)(\mu^2 -\mu^{-2})]y^8$$

$$+[-27(\mu-\mu^{-1}) + 18(\mu^2 -\mu^{-2})]y^{10}$$

$$+[(6735/16)(\mu-\mu^{-1}) - 170(\mu^2-\mu^{-2})+20(\mu^3-\mu^{-3})]y^{12}$$

$$+(1/8)[-16049(\mu-\mu^{-1})+14580(\mu^2-\mu^{-2})-2304(\mu^3-\mu^{-3})]y^{14}$$

$$+ --------\} \qquad (41)$$

Susceptibility per Spin

$$\chi = \lim_{H\to o} [(2m)^2/kT] \{(1/2)(\mu+\mu^{-1})\ y^4$$

$$+[10(\mu+\mu^{-1}) - 5(\mu^2+\mu^{-2})]y^8$$

$$+[-27(\mu+\mu^{-1}) + 36\ (\mu^2+\mu^{-2})]y^{10}$$

$$+[(6735/16)(\mu+\mu^{-1}) -340(\mu^2+\mu^{-2}) + 60\ (\mu^3+\mu^{-3})]y^{12}$$

$$+(1/8)[-16049(\mu+\mu^{-1}) + 29160(\mu^2+\mu^{-2})-6912(\mu^3+\mu^{-3})]y^{14}$$

$$+ \text{-------}\} \tag{42}$$

taking the limit ------

$$\chi_o = [(2m)^2/kT]\ \{y^4+10y^8+18y^{10}+(2255/8)\ y^{12}+(6199/4)y^{14}+\text{---}\} \tag{43}$$

The two spin long range order parameter series expansion is

$$R_2 = 1 - (1/3)[12y^4+96y^6+624y^8+4188y^{10}+(60585/2)y^{12}$$

$$+ 235059\ y^{14}\ \text{-----}] \tag{44}$$

The 4 spin long range order parameter is given by

$$R_4 = 1 - (8/5)(4y^4+32y^6+202y^8+1300y^{10}+(18179/2)y^{12}+69461y^{14}+\text{--}) \tag{45}$$

## 4.2 Series Extrapolations Method

To locate the singularities in the functions whose series have been listed in the previous section, the ratio method of Domb and Sykes[9] and the more recent Padé approximant method have been used. Both methods apply when the terms of the series in question have reached some asymtotic behaviour. In references (5) and (6), Baker has given a detailed account of the applicability of the Padé approximant method. A brief explanation of the two methods follows but for further details the above mentioned references should be consulted.

### 4.2-1 Ratio Method

Assuming an infinite power series has the asymtotic form of a function $(1 - x/x_c)^\gamma$ near some point $x_c$ on the x axis

$$\text{i.e.} \quad (1 - x/x_c)^\gamma \sim \sum_{n=0}^{\infty} a_n x^n \,. \tag{46}$$

Then the limiting form of the ratio of the coefficients is given by

$$\frac{a_{n+1}}{a_n} \sim \frac{1}{x_c}\left[1 - (\gamma/n+1) - (1/n+1)\right]$$

Therefore, if the ratio $(a_{n+1})/(a_n)$ is plotted against $1/(n+1)$ the relation should become linear for sufficiently large n. The intercept on the $(a_{n+1})/(a_n)$ axis will be $1/x_c$ and the limiting slope will be $-(\gamma+1)/(x_c)$.

Figures 4 and 5 show a plot of the ratios of the series for the long range order parameter $R_2$ and the ratios of the specific heat series respectively. From previous studies the long range order parameter is expected to behave asymtotically like $[1-(T/T_c)]^{2\beta}$ near $T_c$ or in terms of y, like $[1-(y^2/y_c{}^2)]^{2\beta}$. $\beta$ is (1/8) for the two dimensional lattices and is approximately (5/16) for three dimensional lattices. By extrapolating the curve of the ratios in Figure 4 our estimates are

$$\frac{kT_c}{J} \simeq 1.72 \quad \pm .12$$

$$\beta = \ .4 \quad \pm .1$$

The specific heat curve singularity at $T_c$ is possibly of the form

$$\frac{Cv}{k} \sim (1 - (T/T_c))^{-1/b} \qquad (47)$$

$$\sim (1 - (y^2/y_c{}^2))^{-1/b}$$

as was indicated for the ferromagnetic f.c.c. and triangular lattices by Domb and Sykes[9]. It is also possible that the

singularity is logarithmically infinite, corresponding to $(1/b) = 0$, on the low temperature side of $T_c$, characteristic of 2 dimensional lattices. If the singularity is logarithmically infinite then the asymtotic form of the ratios of the coefficients of the series $[(d/dy)(C_v/k)]$should be constant.

The ratios are plotted in Figure 5(b) and show a slight increase for large $n$. Again there does not seem to be enough terms to give conclusive evidence of the asymtotic behaviour of the series.

An extrapolation of the ratios of the series of equation (47) failed to give a positive value for 1/b (Figure 5(b)) but the critical point was estimated to be

$$\frac{kT_c}{J} \simeq 1.6 \quad \pm .1$$

### 4.2.2 Padé Approximant Method

If a function is known to possess a singularity of the type $f(x) = (x - x_c)^{-\gamma}$ then its logarithmic derivative will have a simple pole and residue equal to $-\gamma$ at $x_c$

$$\text{i.e.} \quad \frac{d}{dx}[\ln f(x)] = \frac{-\gamma}{x - x_c}$$

In the Padé approximant method the series representing the logarithmic derivative of the function is equated to a ratio of two polynomials $P_M(x)/Q_N(x)$. The numerator is of degree M and the denominator of degree N, where we choose N + M +1 equal to the highest power in our power series. The unknown coefficients of $P_M(x)$ and $Q_N(x)$ are determined by equating coefficients of like powers of x and solving N + M linear equations. The ratio $P_M(x)/Q_N(x)$ is called the [N, M] Padé approximant and we look for the pole of $(d/dx)[\ln f(x)]$ among the N roots of $Q_N(x)$ and $\gamma$ will be found by calculating the residue at the pole.

Long Range Order Approximants

Table III lists values of $T_c$ and $\beta$ from Padé approximant analysis of $(d/du)\ln[R_2]$ where $u = y^2$. The approximants have been listed in Appendix C.

TABLE III

| Padé Approximant | $(kT_c/J)$ | $\beta$ |
|---|---|---|
| [1,1] | 1.85 | .072 |
| [2,2] | 1.81 | .064 |
| [1,3] | 1.85 | .069 |
| [3,1] | - | - |
| [2,3] | 1.93<br>1.57 | .092<br>.0016 |
| [3,2] | 1.725 | .021 |
| [1,4] | 1.84 | .049 |
| [4,1] | - | -* |
| [5,0] | - | - |

PADÉ APPROXIMANT ANALYSIS OF LONG RANGE ORDER SERIES.

* No real roots found near $T_c$.

Specific Heat

In Table IV, estimates of $T_c$ and 1/b were obtained by analysing the logarithmic derivative of $y^{-4}(\log y)^{-2}C_v/k$. The factor $y^4 (\log y)^2$ was removed from the series expansion since it does not appreciably influence the shape of the curve near the sigularity.

TABLE IV

| Padē Approximant | $(kT_c/J)$ | 1/b |
|---|---|---|
| [4,0] | no real roots near Tc | |
| [3,1] | 1.96 | .246 |
| [2,2] | 1.65 | .235 |
| [1,3] | 1.66 | .168 |

PADĒ APPROXIMANT RESULTS OF THE SPECIFIC HEAT SINGULARITY

Magnetic Susceptibility

It is well known that ferromagnetic susceptibilities are infinite at the transition temperature but for anti-ferromagnetic lattices the behaviour is quite different. For the loose packed quadratic and honeycomb lattices, Sykes and Fisher(4) have confirmed that the susceptibility is finite, exhibiting a vertical slope at the transition point and then reaching a maximum slightly above Tc. Baker(5) and Burley(3) have analysed the high temperature antiferromagnetic susceptibility series but their results become uncertain at low temperatures.

It is reasonable to assume then that the magnetic susceptibility of the f.c.c. antiferromagnetic lattice behaves like the loose packed antiferromagnetic lattices near the transition point. If so, we should expect a sigularity (pole) in the derivative of $\chi_0$ and possibly a "zero" above $T_c$. Table V lists the results of a Padé approximant analysis of the derivative of the susceptibility series. The series so far seems quite irregular and ratios of the coefficients do not indicate any asymtotic behaviour. However, a Padé approximant analysis of the derivative of the susceptibility series was attempted using two approximants. The results of the

calculations are listed below in Table V. The [1,4] approximant gave a somewhat higher answer for $kT_c/J$ than previous estimates and the [2,3] approximant failed completely.

TABLE V

| Padé Approximant | $kT_c/J$ |
|---|---|
| [1,4] | 2.15 |
| [2,3] | - |

PADÉ APPROXIMANT ANALYSIS OF THE SUSCEPTIBILITY SERIES

## 4.3 Critical Entropy

The entropy can be calculated using equation (15) and directly integrating the specific heat curve. It is of interest to know the critical entropy given by

$$S_c = \int_0^{T_c} \frac{C_v}{T} \, dT \tag{48}$$

For all lattices $S_\infty$ is $k \ln 2 = 0.6931\ k$. The percentage increase of the entropy to $T_c$ increases with coordination number. For the f.c.c. ferromagnetic lattice ($q = 12$) $S_c/k = 0.597$ so that a very large portion of the entropy change takes place below $T_c$. For the two dimensional quadratic lattice $S_c/k = 0.306$ is much less. Our estimate for the f.c.c. antiferromagnetic lattice is $S_c/k = 0.312$ which is remarkably near to the value for the quadratic lattice. This is strong evidence that the effective dimensionality of the f.c.c. antiferromagnetic lattice is two rather than three.

The integral in equation (48) was obtained by fitting the specific heat series to a logarithmic singularity at $T_c$, similar to a method used by Wakefield[12].

We equate the specific heat series to a logarithmically infinite function as follows.

$$\frac{C_v}{k} = [\log y]^2 \left\{ 16y^4 + 144y^6 + 960y^8 + 6850y^{10} + 54102y^{12} + (941339/2)y^{14} + \text{------} \right\}$$

$$= A(\ln y)^2 \ln (1 - (y^2/y_c^{\,2})) + (\ln y)^2 \sum_{n=o}^{7} a_{2n} y^{2n} \qquad (49)$$

The first term on the right side of (49) represents the singularity and the polynomial ensures that function well represents the specific heat for values of y much less than $y_c$. We chose $y_c^2$ = .1307 from the [1,3] Padé Approximant in Table IV. Equating coefficients of equal powers of y we end up with eight equations and nine unknowns, A, $a_o$, $a_2$ --- $a_{14}$. However, we can set $a_{14}$ equal to zero and solve for the remaining eight unknown coefficients. Setting $a_{14}$ to zero is effectively saying that our series has settled down to asymtotic behaviour. The new series obtained in this way is:

$$\frac{C_v}{k} = 2.13802\ (\ln y)^2 \ln [1 - (y^2/(.13069)^2)]$$
$$- (\ln y)^2\ \{16.35932\ y^2 + 46.58828\ y^4 + 175.2723\ y^6 + 872.238\ y^8 + 4365.785\ y^{10} + 17414.454\ y^{12}\} \qquad (50)$$

In terms of the variable $\tau = T/T_c$, the critical entropy is given by

$$\frac{S_c}{k} = \int_0^1 \frac{(C_v/k)}{\tau}\, d\tau$$

In Figure 6 $(C_v/k\tau)$ is plotted against $\tau$ using equation (50). Thus the critical entropy is given by the area under the curve and we find $S_c/k = .312$ for the f.c.c.

We have also plotted the specific heat curves for the antiferromagnetic quadratic lattice and the ferromagnetic f.c.c. lattice for comparison. The area under the curves for the f.c.c. antiferromagnet and the simple quadratic lattice are almost identical (.312 and .306 respectively) but their shapes differ significantly. We see also that a much greater percentage of the entropy change occurs below $T_c$ for the f.c.c. ferromagnet than for the corresponding antiferromagnet.

APPLICATION OF RATIO METHOD TO LONG RANGE ORDER SERIES

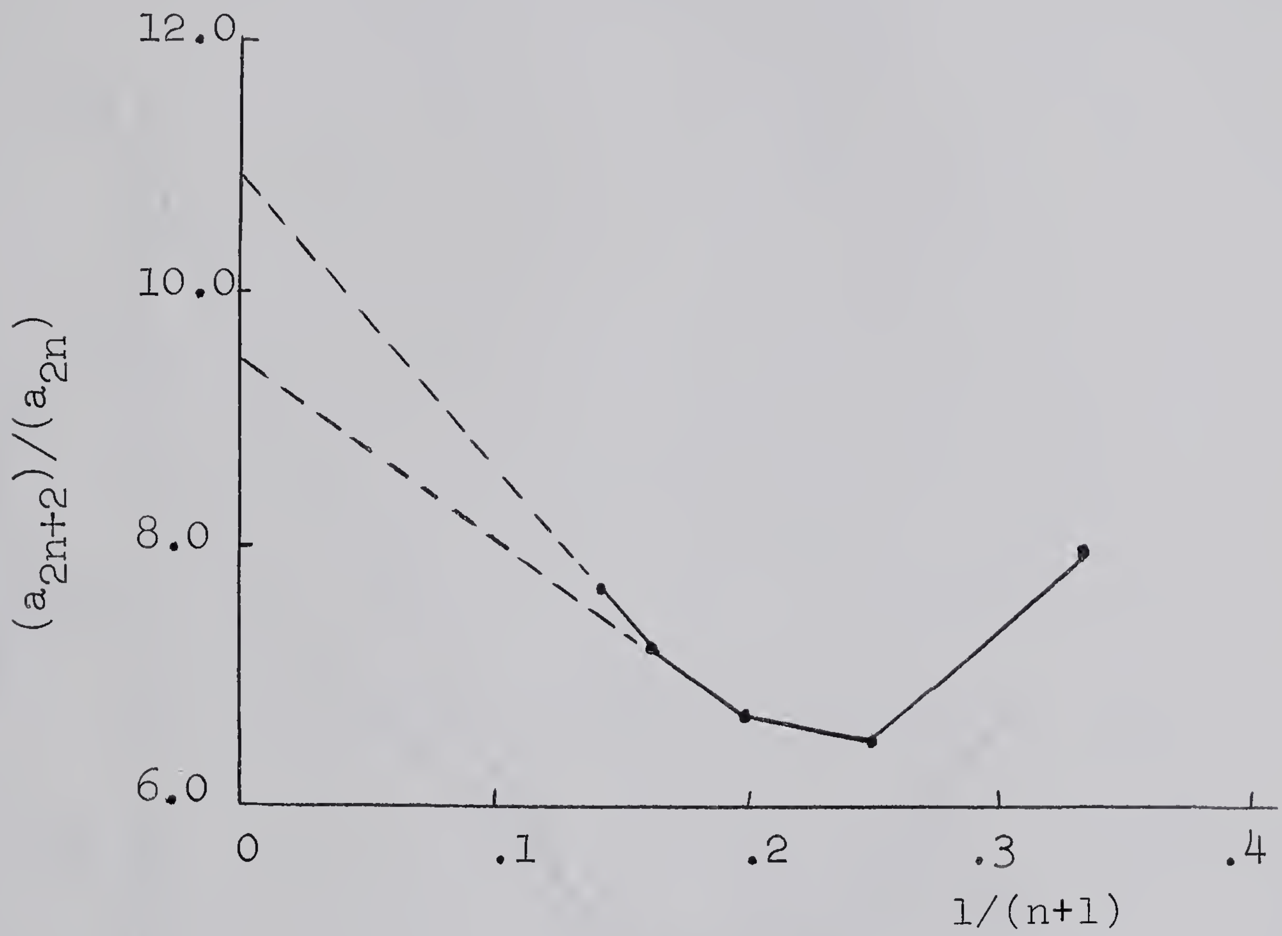



Figure 4

The ratios $(a_{2n+2})/(a_{2n})$ are plotted against 1/n

where $\sum_{a=0}^{7} a_{2n}\, y^{2n} = R_2 = 1 - 4y^4 - 32y^6 - 208y^8$

$$- 1396y^{10} - (20195/2)y^{12} - 78353y^{14}$$

APPLICATION OF RATIO METHOD TO SPECIFIC HEAT SERIES

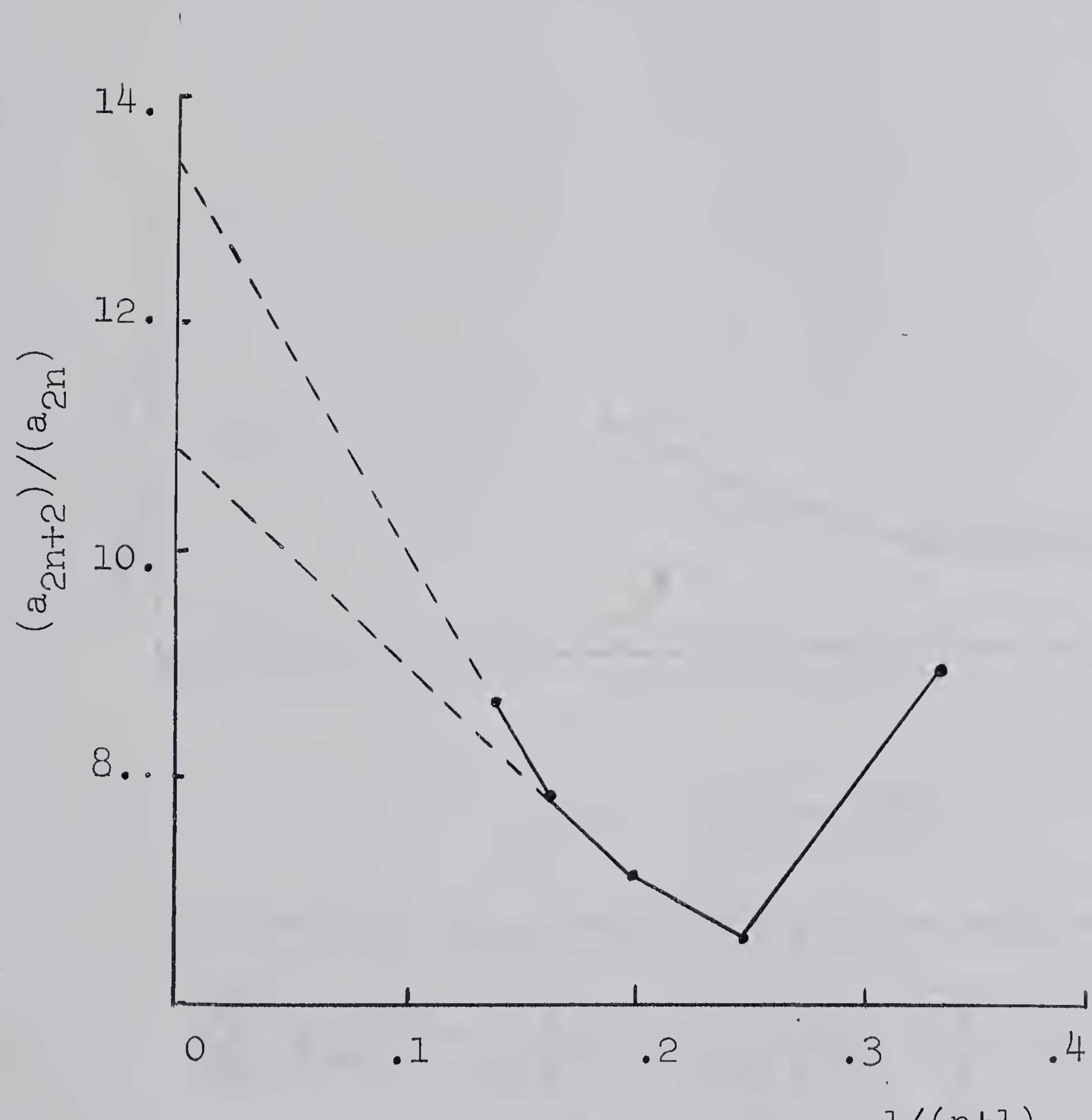



Figure 5(a)

The ratios $(a_{2n+2})/(a_{2n})$ are plotted against 1/n

where $\sum_{n=2}^{7} a_{2n}\, y^{2n} = 16y^4 + 144y^6 + 960y^8 + 6880y^{10} + 54102y^{12} + (941339/2)y^{14}$

APPLICATION OF RATIO METHOD TO THE DERIVATIVE OF THE SPECIFIC HEAT SERIES

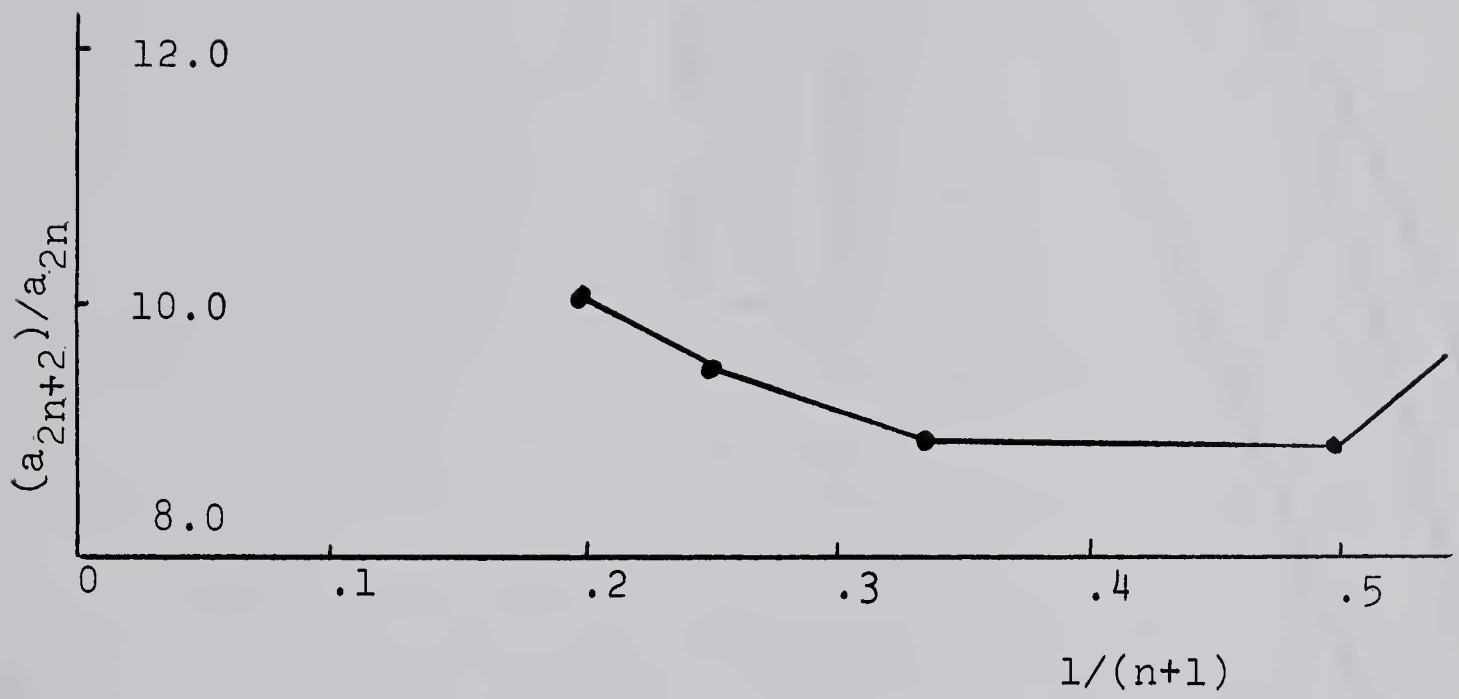


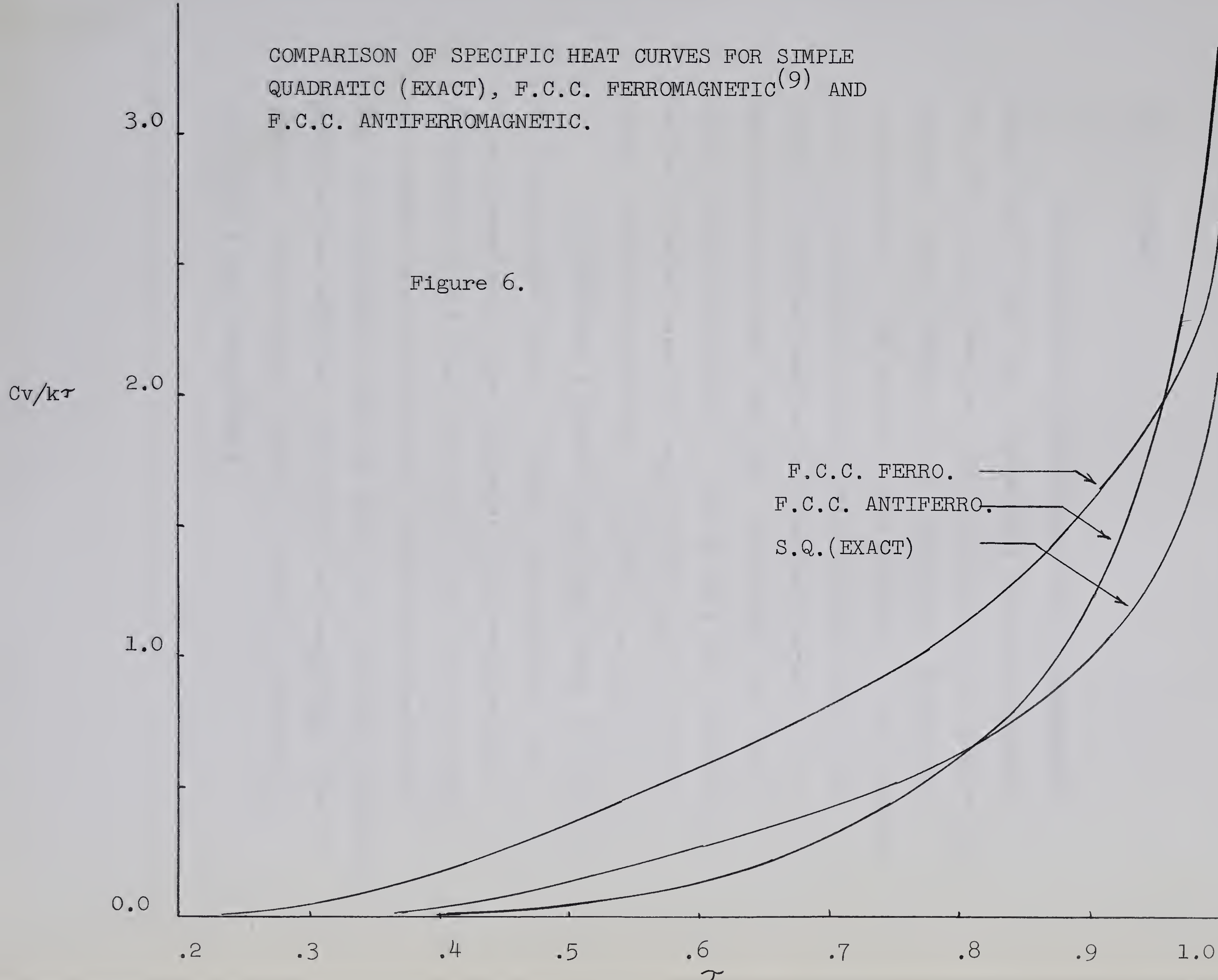

Figure 5(b)

The ratios $(a_{2n+2}/a_2n)$ are plotted against $1/(n+1)$ where

$$\sum_{n=1}^{6} a_{2n}y^{2n} = \frac{d}{d(y^2)} \left[ \frac{1}{(\log y)^2} \quad \frac{C_v}{k} \right]$$

$$= 32y^2 + 432y^4 + 3480y^6 + 34{,}250y^8$$

$$= 324612y^{10} + 3294686.5y^{12}$$




COMPARISON OF SPECIFIC HEAT CURVES FOR SIMPLE QUADRATIC (EXACT), F.C.C. FERROMAGNETIC[9] AND F.C.C. ANTIFERROMAGNETIC.

Figure 6.

# 5. Summary and Discussion

## 5.1 Summary

The problem of determining the low temperature properties of the f.c.c. Ising model antiferromagnetic lattice has long been an outstanding problem in theoretical physics, mostly because of the complicated ground state configuration. The only feasible approach to the problem is by configuration counting methods which quickly becomes an enormous task as more and more terms in the expansion series are required. Previously only three terms had been calculated[8] but we have now extended the series to seven terms and many of the contributions to higher terms have also been calculated (see Appendix A).

An extensive study of long range order in the f.c.c. lattice was also made. A series representation for a three dimensional order parameter which measures the correlation among four spins at the vertices of a large tetrahedron was calculated and a relation between this four spin correlation and the more usual two spin correlation parameter was obtained. To obtain the long range order expansion series special counting theorems were developed to simplify the calculations.

The transition temperature Tc is characterized by the disappearance of long range order. Using the Padé approximant method[5,6] and the ratio method[9] we have estimated that the long range order disappears at

$$\frac{kTc}{J} = 1.9 \pm .1$$

From the low temperature expansion series of the partition function in a magnetic field H, series of many other thermodynamic functions have been calculated including the internal energy U, the specific heat $C_v$, the entropy S, the magnetization M and the zero field susceptibility $\chi_o$.

Since the specific heat series was somewhat regular in comparison to the series for the other thermodynamic functions, and also because it is expected to exhibit a weak singularity at the transition point, our studies were concentrated on its properties.

In general the Padé approximant and ratio method of series extrapolation confirmed the existence of a transition point near $(kT_c/J) = 1.9$ and although there is still doubt, the singularity in $C_v$ seems to be logarithmic in nature on the low temperature side of $T_c$.

The specific heat series was fitted to a logarithmic singularity below $T_c$ and compared to the specific heat curves of the two dimensional quadratic and the f.c.c. ferromagnetic lattices (Figure 6). It was interesting that the curve for the f.c.c. antiferromagnet more closely resembled the curve for the quadratic lattice than that of the f.c.c. ferromagnet. Possible reasons for this are discussed in the next section.

The critical entropy $S_c$ was calculated by direct integration of the specific heat curve.

$$\text{i.e.} \quad S_c = \int_0^{T_c} (C_V/T)dT$$

The estimate for $S_c$ was 0.312k as compared to 0.306k for the simple quadratic lattice and 0.597k for the f.c.c. anti-ferromagnetic lattice. The implications here are discussed in the next section.

The susceptibility series was too irregular to attempt an extrapolation. It appears at least two more terms will be required before the behaviour near $T_c$ can be determined.

For low temperature in the range $T/T_c < .5$ all the series expansions appear to converge rapidly and therefore the thermodynamic functions are well represented.

by the first few terms of their associated series. But in general it was difficult to determine the asymtotic form of the series because of this insufficient number of terms. However, the graphs of the coefficient ratios of the specific heat and the long range order series indicate that the coefficients may be approaching asymtotic behaviour and if this is the case, two or three more terms should be sufficient to get some conclusive answers.

## 5.2 Effective Dimensionality.

The ground state has been shown to consist of layers of antiferromagnetically ordered quadratic nets in which the spin states do not seem to influence the spin states in adjacent ordered layers, at least for very low temperatures. Because of this unusual ordering in the ground state the question naturally arose as to whether the thermodynamic properties would be characteristic of the two dimensional quadratic antiferromagnetic lattice or would they more closely resemble the properties of the three dimensional close packed ferromagnetic lattices.

The answer is probably something in between. From the point of view of order among the spins in the lattice sites we have shown that order is essentially only in two dimensions. But we can still define three dimensional order parameters which correlate two and four spins at arbitrary locations on the lattice as long as they lie along a straight line of odd number of bond lengths for two spins and are at the vertices of a tetrahedron for four spins. The specific heat curves (figure 6) show that there is a remarkable difference between the specific heat of f.c.c. antiferromagnet and the f.c.c. ferromagnet; a much larger portion of the curve for f.c.c. ferromagnet being below Tc. However the f.c.c. anti-ferromagnetic curve is similar to that of the simple quadratic

except for the difference in area distribution. This difference must somehow be related to the way in which the quadratic layers of the f.c.c. antiferromagnetic lattice interact, otherwise the curves would be identical in nature.

Our estimate is $S_c = .312k$ showing that about 45% of the total entopy change takes place below $T_c$. For the ferromagnetic and loose packed lattices $S_c$ is roughly proportional to q. $S_c = .306k$ for the simple quadratic (q = 4) and $S_c = .597k$ for the f.c.c. ferromagnetic lattice (q = 12). However our estimate for the f.c.c. antiferromagnet, being much lower than the critical entropy for the f.c.c. ferromagnet seems to indicate that it is not really q that decides the magnitude of $S_c$ but possible an "effective q" where in general the "effective q" would be less than q for close packed antiferromagnetic lattices and equal to q for ferromagnetic and antiferromagnetic loose packed lattices.

## 5.3 Comments on Experimental Measurements

On past occasions, the Ising model has produced reasonable good qualitative agreement with experiment and we should expect the f.c.c. antiferromagnet lattice to be no exception. There are many substances in nature which have a f.c.c. antiferromagnetic structure. Among them are MnO, MnS, FeO, CoO, NiO, and the salts $K_2IrCl_6$ and $(NH_4)_2IrCl_6$. MnO has been studied by Bizette* et al and exhibits a logarithmic type singularity in the specific heat. The susceptibility reaches a maximum at the critical point.

Domb and Miedema[(13)] have written an excellent summary of experimental measurements of transitions in solids which include a table** of thermodynamic properties of antiferromagnetic salts with loose packed structures. For the cases of spin 1/2, the values of $S_c/k$ are smaller than the corresponding close packed antiferromagnetic values which seem to agree with our result.

* See reference (15), p 436 for graphs of MnO properties

** See Table 9, p 340 of reference (13)

## 5.4 Further Studies.

There are some properties of the f.c.c. spin 1/2 Ising model which have not been studied in this thesis but are still of considerable interest to theoretical physicists. There is the calculation of middle range order parameters (between second, third etc., nearest neighbours). The short range order, between nearest neighbours is linearly related to the internal energy and therefore has essentially been calculated. A study of all these parameters would more clearly specify the manner in which order spreads as temperature is decreased to $0^{\circ}$.

The critical energy would also be of interest since this quantity can be compared to experimental measurements.

Very little can be said about the susceptibility series at present because of irregularity in the first few terms of the series. Therefore more terms will be required before information about the behaviour near the transition temperature and especially about the maximum above $T_c$ can be obtained.

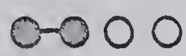
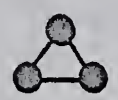
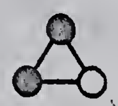
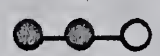
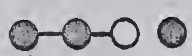
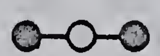
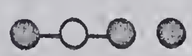
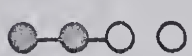
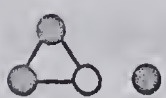
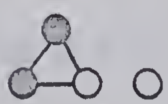
# APPENDIX A

## Face Centered Cubic Antiferromagnetic Low Temperature Lattice Configurations *

*Notation: The superscript of "C" is half the energy associated with overturning the spins. The subscript numbers the configurations and specifies the number of vertices in the graph. Thus, $C^{22}_{5096}$ is associated with energy 44J, has five vertices and is the 96th five spin graph.

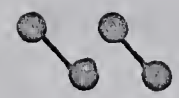
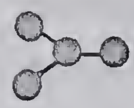
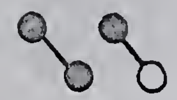
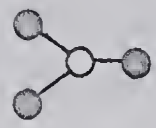
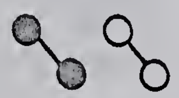
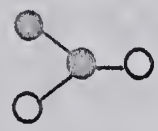
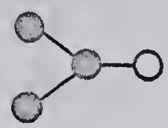
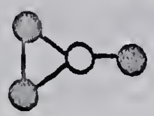
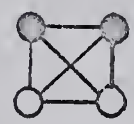
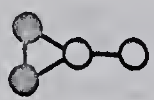
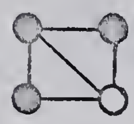
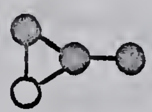
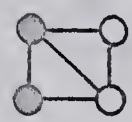
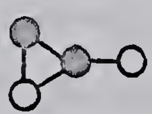
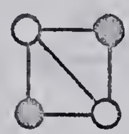
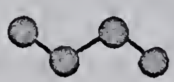
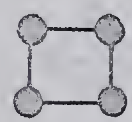
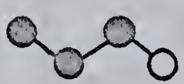
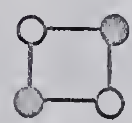
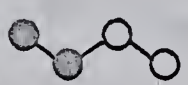
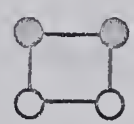
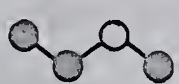
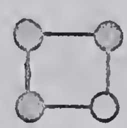
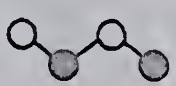
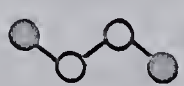
Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

1, 2, 3, and 4 Vertices

| | | | | | |
|---|---|---|---|---|---|
| $C_{11}^{4}$ |  | $\frac{1}{2}$ | $C_{310}^{12}$ |  | $\frac{1}{16}(N^2-42N+480)$ |
| $C_{21}^{8}$ |  | $\frac{1}{8}(N-10)$ | $C_{401}^{18}$ |  | $\frac{1}{8}(N^2-42N+480)$ |
| $C_{22}^{8}$ |  | $\frac{1}{4}(N-16)$ | $C_{402}^{18}$ |  | $\frac{1}{8}(2N^2-112N+1720)$ |
| $C_{23}^{10}$ |  | 1 | $C_{403}^{18}$ |  | $\frac{1}{8}(N^2-58N+896)$ |
| $C_{24}^{6}$ |  | 4 | $C_{404}^{14}$ |  | $\frac{1}{8}(4N^2-200N+2712)$ |
| $C_{31}^{24}$ |  | 0 | $C_{405}^{14}$ |  | $N^2-56N+870$ |
| $C_{32}^{10}$ |  | 4 | $C_{406}^{20}$ |  | $\frac{1}{2}(3N-64)$ |
| $C_{33}^{16}$ |  | 3 | $C_{407}^{20}$ |  | $\frac{1}{2}(3N-95)$ |
| $C_{34}^{12}$ |  | 8 | $C_{408}^{16}$ |  | $4N-103$ |
| $C_{35}^{8}$ |  | 10 | $C_{409}^{12}$ |  | $\frac{1}{2}(10N-239)$ |
| $C_{36}^{14}$ |  | $\frac{1}{2}N-8$ | $C_{410}^{16}$ |  | $4N-112$ |
| $C_{37}^{14}$ |  | $\frac{1}{2}N-12$ | $C_{411}^{12}$ |  | $5N-147$ |
| $C_{38}^{10}$ |  | $\frac{1}{2}N-40$ | $C_{412}^{14}$ |  | $2N-48$ |
| $C_{39}^{12}$ |  | $\frac{1}{48}(N^2-30N+248)$ | $C_{413}^{14}$ |  | $2N-52$ |

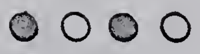
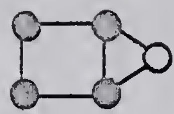
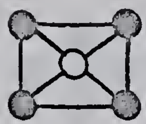
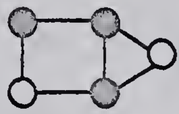
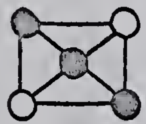
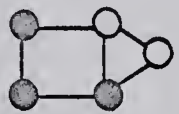
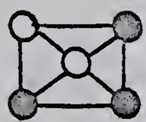
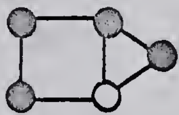
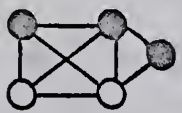
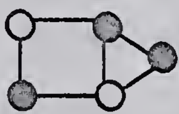
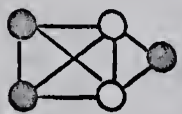
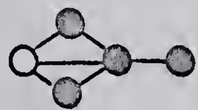
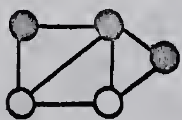
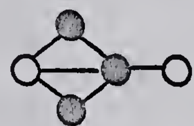
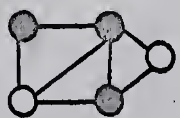
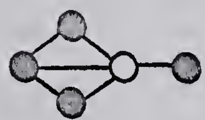
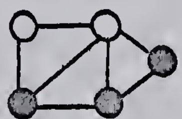
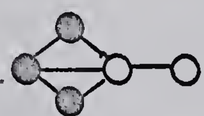
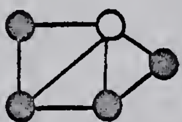
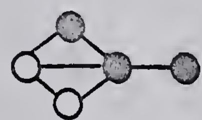
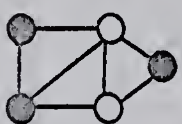
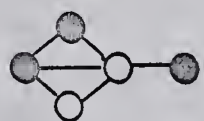
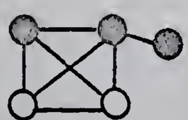
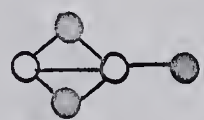
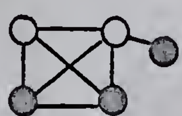
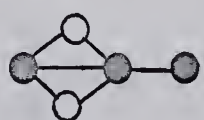
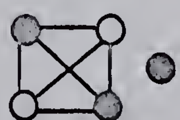
# Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

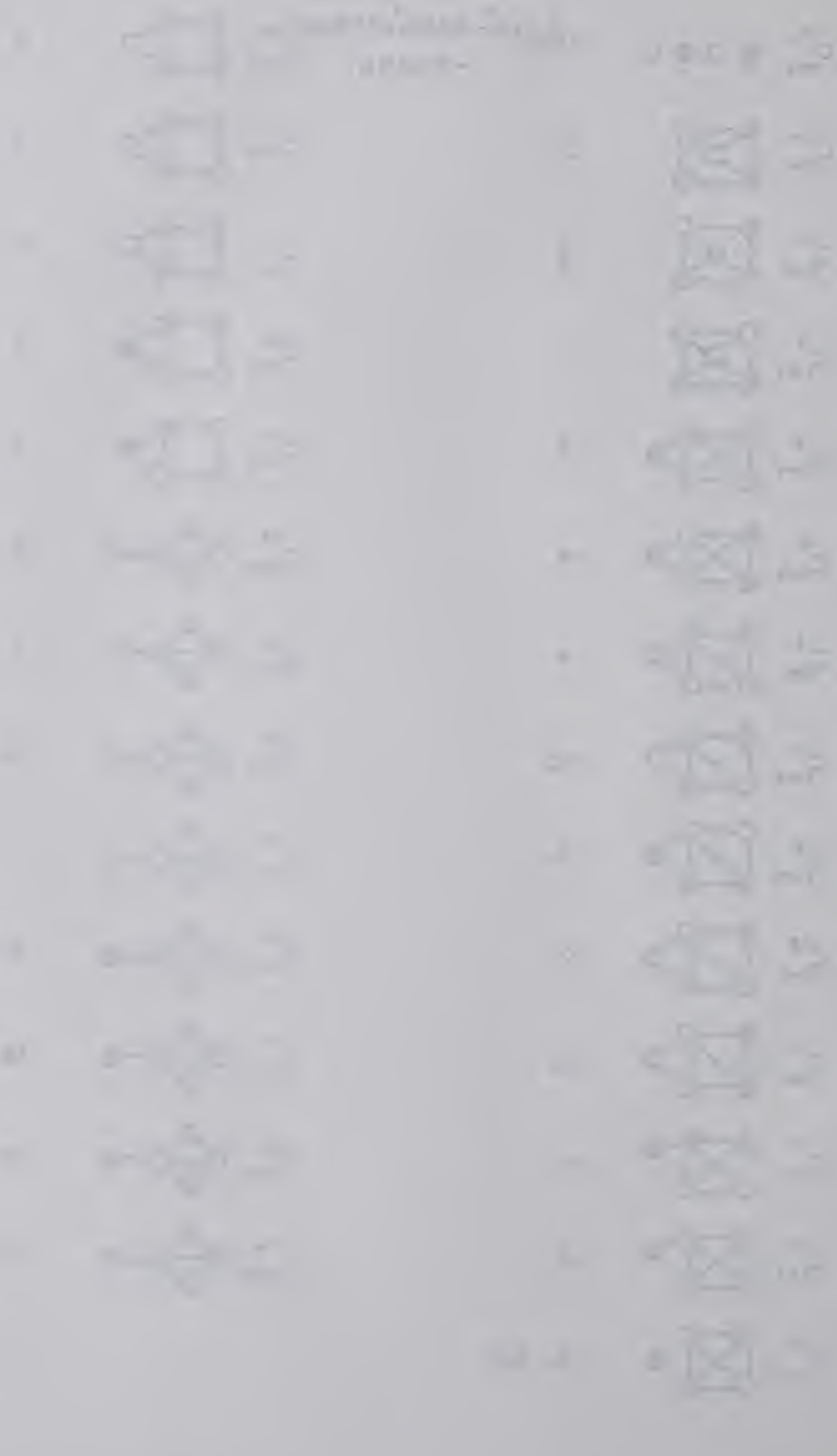
## 4 Vertices

| | | | | | |
|---|---|---|---|---|---|
| $C_{414}^{20}$ |  | $\frac{1}{2}N - 12$ | $C_{428}^{22}$ |  | 2 |
| $C_{415}^{16}$ |  | $4N - 119$ | $C_{429}^{10}$ |  | 8 |
| $C_{416}^{20}$ |  | $N - 34$ | $C_{430}^{14}$ |  | 8 |
| $C_{417}^{12}$ |  | $8N - 235$ | $C_{431}^{18}$ |  | 4 |
| $C_{418}^{12}$ |  | 16 | $C_{432}^{12}$ |  | 2 |
| $C_{419}^{16}$ |  | 4 | $C_{433}^{14}$ |  | 4 |
| $C_{420}^{16}$ |  | 16 | $C_{434}^{14}$ |  | 8 |
| $C_{421}^{12}$ |  | 24 | $C_{435}^{10}$ |  | 4 |
| $C_{422}^{22}$ |  | 8 | $C_{436}^{24}$ |  | $\frac{1}{4}$ |
| $C_{423}^{18}$ |  | 23 | $C_{437}^{8}$ |  | $\frac{3}{2}$ |
| $C_{424}^{18}$ |  | 16 | $C_{438}^{-}$ |  | 0 |
| $C_{425}^{14}$ |  | 39 | $C_{439}^{16}$ |  | $\frac{1}{2}$ |
| $C_{426}^{10}$ |  | 94 | $C_{440}^{16}$ |  | $\frac{1}{384}(N^3 - 60N^2 + 1292N - 10128)$ |
| $C_{427}^{14}$ |  | 16 | $C_{441}^{16}$ |  | $\frac{1}{384}(4N^3 - 312N^2 + 8672 - 87360)$ |

# Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

## 5 Vertices

| | | | | | |
|---|---|---|---|---|---|
| $C^{16}_{442}$ |  | $\frac{1}{192}(3N^3-252N^2+7596N-83808)$ | $C^{24}_{5015}$ |  | 2 |
| $C^{20}_{5001}$ |  | $\frac{1}{2}$ | $C^{16}_{5016}$ |  | 2 |
| $C^{12}_{5002}$ |  | $\frac{3}{2}$ | $C^{-}_{5017}$ |  | 0 |
| $C^{16}_{5003}$ |  | 1 | $C^{20}_{5018}$ |  | 2 |
| $C^{18}_{5004}$ |  | 8 | $C^{12}_{5019}$ |  | 6 |
| $C^{12}_{5006}$ |  | 4 | $C^{20}_{5020}$ |  | 8 |
| $C^{18}_{5007}$ |  | 4 | $C^{16}_{5021}$ |  | 7 |
| $C^{14}_{5008}$ |  | 10 | $C^{16}_{5022}$ |  | 14 |
| $C^{18}_{5009}$ |  | 6 | $C^{20}_{5023}$ |  | 1 |
| $C^{18}_{5010}$ |  | 2 | $C^{20}_{5024}$ |  | 8 |
| $C^{14}_{5011}$ |  | 14 | $C^{16}_{5025}$ |  | 18 |
| $C^{18}_{5012}$ |  | 4 | $C^{12}_{5026}$ |  | 17 |
| $C^{14}_{5013}$ |  | 8 | $C^{16}_{5027}$ |  | 11 |
| $C^{16}_{5014}$ |  | N - 28 | | | |



## Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

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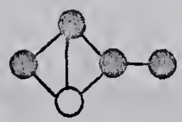
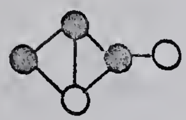
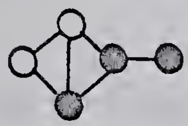
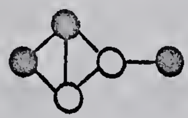
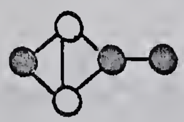
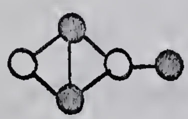
$C^{20}_{5028}$  16

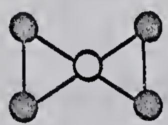
$C^{16}_{5029}$  24

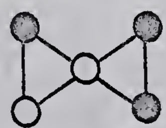
$C^{20}_{5030}$  16

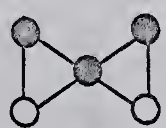
$C^{16}_{5031}$  24

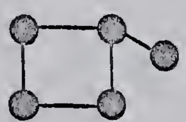
$C^{16}_{5032}$  8

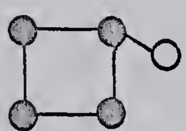
$C^{12}_{5033}$  32

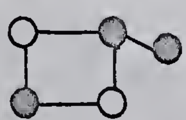
$C^{16}_{5034}$  7

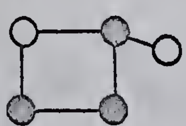
$C^{16}_{5035}$  10

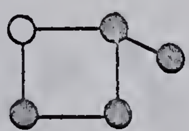
$C^{16}_{5036}$  19

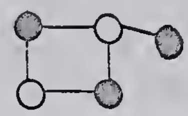
$C^{30}_{5037}$  2

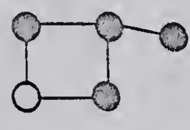
$C^{26}_{5038}$  2

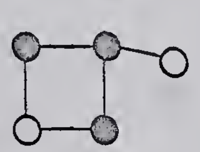
$C^{14}_{5039}$  3

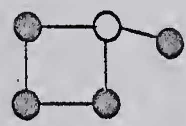
$C^{18}_{5040}$  3

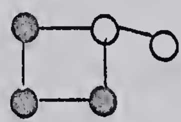
$C^{22}_{5041}$  1

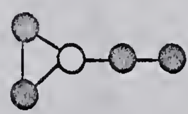
$C^{10}_{5042}$  9

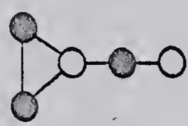
$C^{22}_{5043}$  1

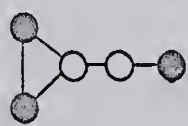
$C^{18}_{5044}$  1

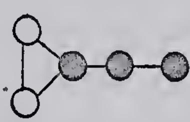
$C^{18}_{5045}$  1

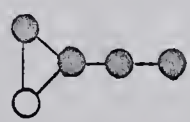
$C^{22}_{5046}$  1

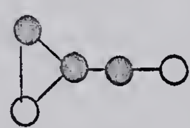
$C^{18}_{5047}$  9

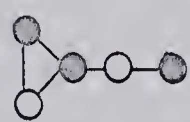
$C^{14}_{5048}$  7

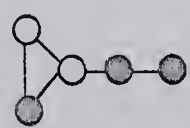
$C^{18}_{5049}$  9

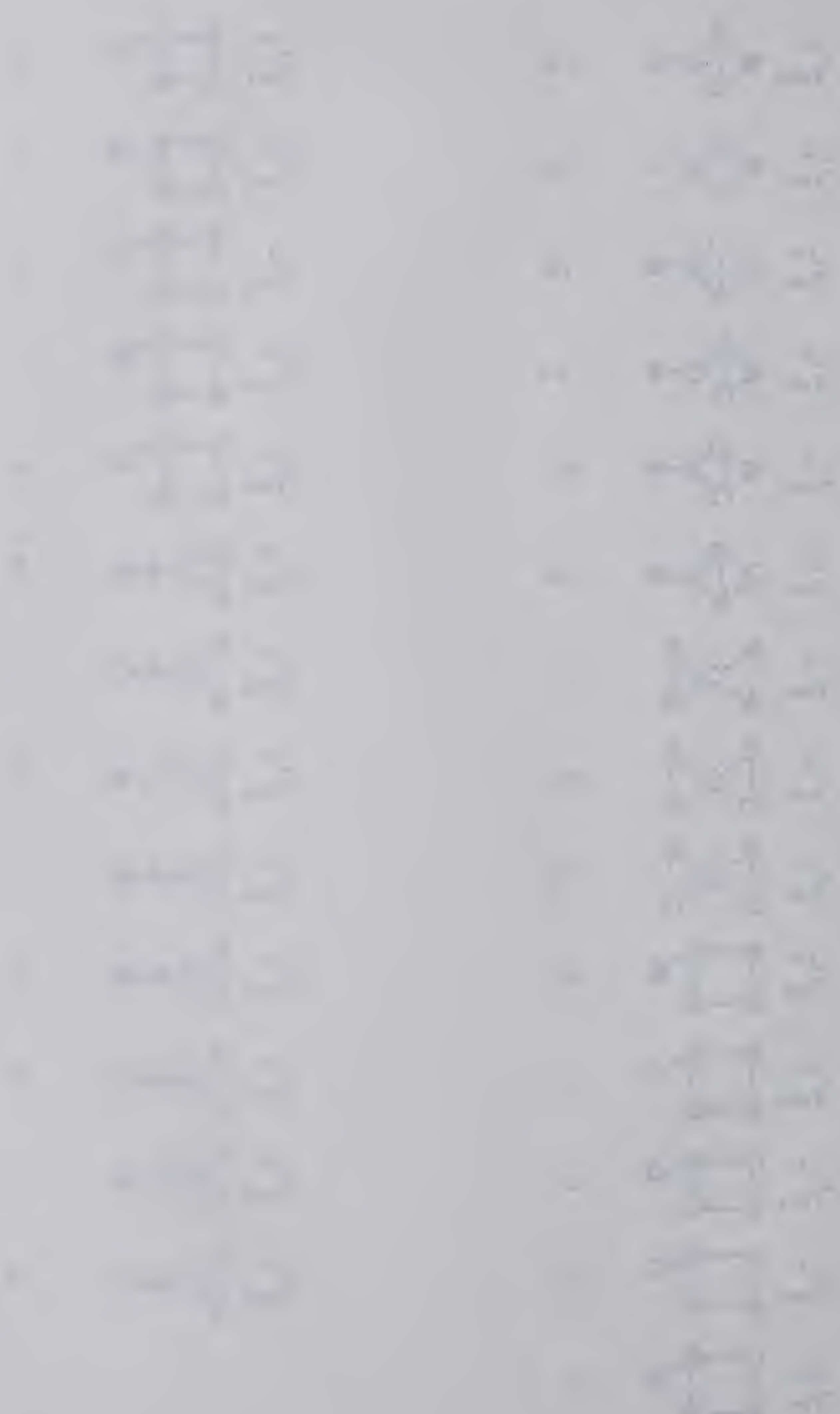
$C^{22}_{5050}$  11

$C^{22}_{5051}$  11

$C^{18}_{5052}$  9

$C^{14}_{5053}$  7

$C^{18}_{5054}$  9



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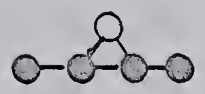
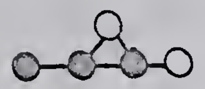
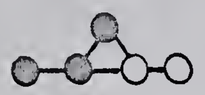
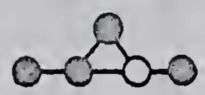
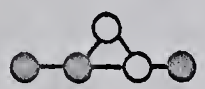
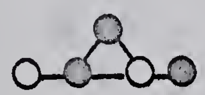
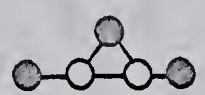
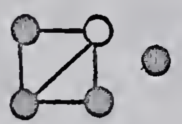
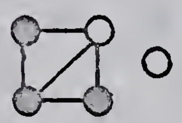
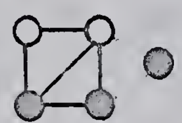
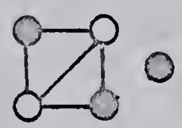
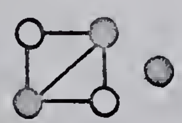
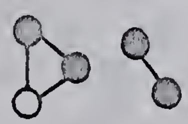
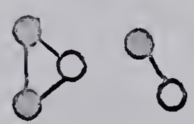
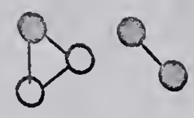
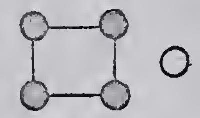
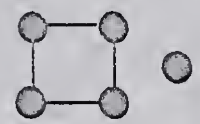
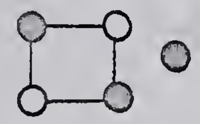
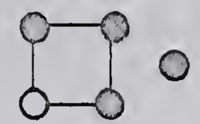
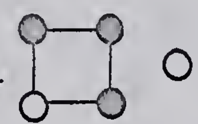
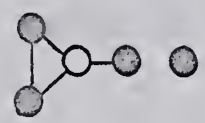
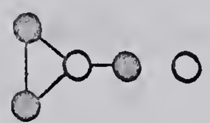
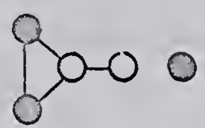
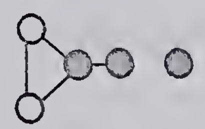
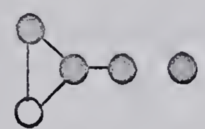
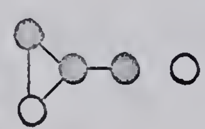
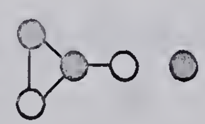
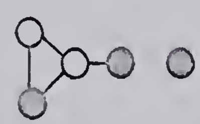
| $C^{22}_{5055}$ |  | 14 |
| $C^{18}_{5056}$ |  | 46 |
| $C^{22}_{5057}$ |  | 16 |
| $C^{18}_{5058}$ |  | 62 |
| $C^{18}_{5059}$ |  | 24 |
| $C^{14}_{5060}$ |  | 90 |
| $C^{14}_{5061}$ |  | 36 |
| $C^{18}_{5062}$ |  | $2N-56$ |
| $C^{18}_{5063}$ |  | $2N-64$ |
| $C^{18}_{5064}$ |  | $4N-120$ |
| $C^{14}_{5065}$ |  | $2N-56$ |
| $C^{14}_{5066}$ |  | $2N-64$ |
| $C^{20}_{5067}$ |  | $4N-140$ |
| $C^{16}_{5068}$ |  | $16N-576$ |
| $C^{20}_{5069}$ |  | $4N-148$ |
| $C^{28}_{5070}$ |  | $\frac{1}{8}(N-36)$ |
| $C^{28}_{5071}$ |  | $\frac{1}{8}(N-24)$ |
| $C^{12}_{5072}$ |  | $\frac{1}{4}(3N-90)$ |
| $C^{20}_{5073}$ |  | $\frac{1}{4}(N-26)$ |
| $C^{20}_{5074}$ |  | $\frac{1}{4}(N-34)$ |
| $C^{16}_{5075}$ |  | $8N-222$ |
| $C^{16}_{5076}$ |  | $8N-282$ |
| $C^{20}_{5077}$ |  | $2N-64$ |
| $C^{20}_{5078}$ |  | $2N-64$ |
| $C^{20}_{5079}$ |  | $8N-234$ |
| $C^{20}_{5080}$ |  | $8N-270$ |
| $C^{16}_{5081}$ |  | $12N-400$ |
| $C^{16}_{5082}$ |  | $12N-360$ |

Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS
5 Vertices

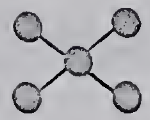
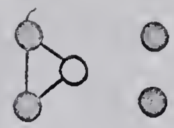
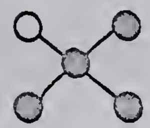
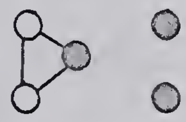
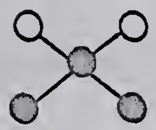
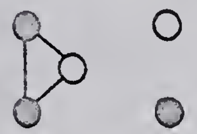
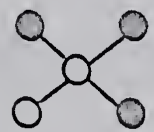
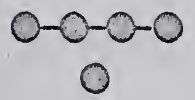
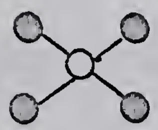
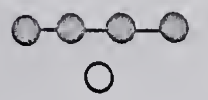
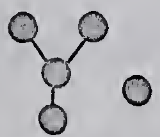
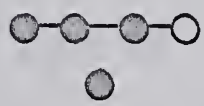
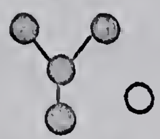
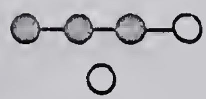
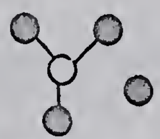
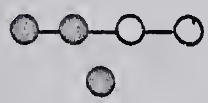
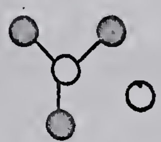
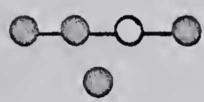
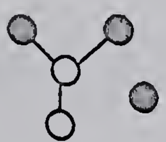
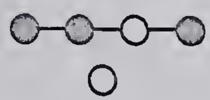
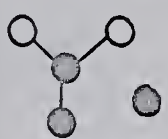
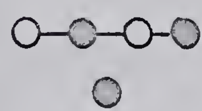
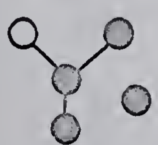
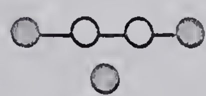
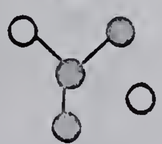
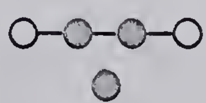
| | | | | | |
|---|---|---|---|---|---|
| $C^{28}_{5111}$ |  | $\frac{1}{2}$ | $C^{18}_{5124}$ |  | $\frac{1}{2}(N^2-58N+904)$ |
| $C'_{5112}$ |  | 0 | $C^{18}_{5125}$ |  | $\frac{1}{2}(N^2-62N+1024)$ |
| $C^{20}_{5113}$ |  | $\frac{3}{2}$ | $C^{18}_{5126}$ |  | $N^2-66N+1200$ |
| $C^{16}_{5114}$ |  | 1 | $C^{26}_{5127}$ |  | $4N-108$ |
| $C^{12}_{5115}$ |  | $\frac{3}{2}$ | $C^{26}_{5128}$ |  | $4N-157$ |
| $C^{26}_{5116}$ |  | $N-26$ | $C^{22}_{5129}$ |  | $\frac{23}{2}N-358$ |
| $C^{26}_{5117}$ |  | N 39 | $C^{22}_{5130}$ |  | $\frac{23}{2}N-411$ |
| $C^{14}_{5118}$ |  | $4N-111$ | $C^{22}_{5131}$ |  | $8N-270$ |
| $C^{14}_{5119}$ |  | $4N-151$ | $C^{18}_{5132}$ |  | $\frac{39}{2}N-580$ |
| $C^{18}_{5120}$ |  | $4N-127$ | $C^{18}_{5133}$ |  | $\frac{39}{2}N-729$ |
| $C^{18}_{5121}$ |  | $4N-138$ | $C^{14}_{5134}$ |  | $47N-1569$ |
| $C^{22}_{5122}$ |  | $2N-61$ | $C^{18}_{5135}$ |  | $8N-256$ |
| $C^{22}_{5123}$ |  | $2N-71$ | $C^{18}_{5136}$ |  | $8N-284$ |

Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS
5 Vertices

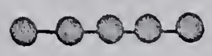
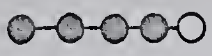
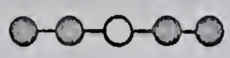
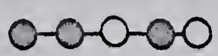
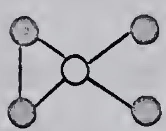
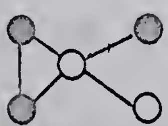
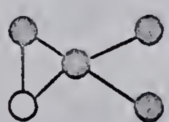
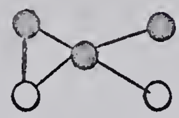
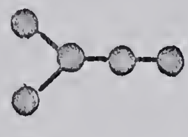
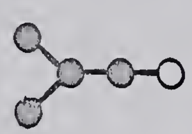
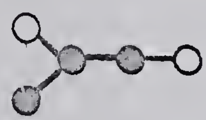
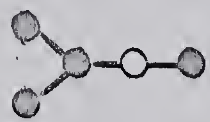
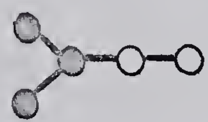
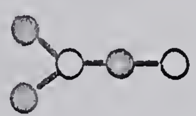
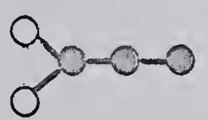
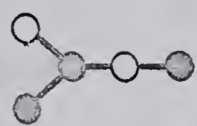
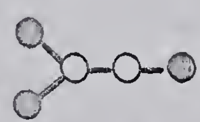
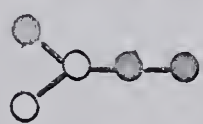
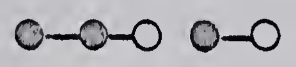
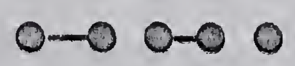
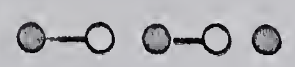
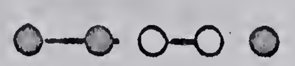
| | | |
|---|---|---|
| $C^{28}_{5083}$ |  | 22 |
| $C^{24}_{5084}$ |  | 61 |
| $C^{20}_{5085}$ |  | 113 |
| $C^{20}_{5086}$ |  | $\frac{77}{2}$ |
| $C^{24}_{5087}$ |  | 46 |
| $C^{16}_{5088}$ |  | 183 |
| $C^{16}_{5089}$ |  | 156 |
| $C^{20}_{5090}$ |  | $\frac{89}{2}$ |
| $C^{20}_{5091}$ |  | 64 |
| $C^{12}_{5092}$ |  | 223 |
| $C^{14}_{5093}$ |  | 10 |
| $C^{18}_{5094}$ |  | 6 |
| $C^{14}_{5095}$ |  | 14 |
| $C^{22}_{5096}$ |  | 8 |
| $C^{18}_{5097}$ |  | 10 |
| $C^{28}_{5098}$ |  | 14 |
| $C^{24}_{5099}$ |  | 22 |
| $C^{24}_{5100}$ |  | 19 |
| $C^{20}_{5101}$ |  | 30 |
| $C^{20}_{5102}$ |  | 19 |
| $C^{24}_{5103}$ |  | 8 |
| $C^{16}_{5104}$ |  | 46 |
| $C^{12}_{5106}$ |  | 102 |
| $C^{20}_{5107}$ |  | 21 |
| $C^{16}_{5108}$ |  | 71 |
| $C^{16}_{5109}$ |  | 32 |
| $C^{20}_{5110}$ |  | 30 |

Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

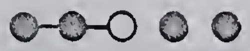
5 Vertices

| | | |
|---|---|---|
| $C^{26}_{5137}$ |  | $3N-94$ |
| $C^{26}_{5138}$ |  | $3N-131$ |
| $C^{22}_{5139}$ |  | $12N-465$ |
| $C^{22}_{5140}$ |  | $8N-299$ |
| $C^{18}_{5141}$ |  | $32N-1247$ |
| $C^{22}_{5142}$ |  | $8N-318$ |
| $C^{18}_{5143}$ |  | $10N-354$ |
| $C^{14}_{5144}$ |  | $40N-1532$ |
| $C^{18}_{5145}$ |  | $10N-426$ |
| $C^{24}_{5146}$ |  | $\frac{1}{4}(N^2-56N+856)$ |
| $C^{24}_{5147}$ |  | $\frac{1}{4}(N^2-72N+1454)$ |
| $C^{20}_{5148}$ |  | $\frac{1}{2}(4N^2-263N+4716)$ |
| $C^{20}_{5149}$ |  | $\frac{1}{2}(4N^2-295N+6026)$ |
| $C^{16}_{5150}$ |  | $\frac{1}{2}(8N^2-555N+10633)$ |
| $C^{24}_{5151}$ |  | $\frac{1}{2}(N^2-74N+1476)$ |
| $C^{22}_{5152}$ |  | $\frac{1}{48}(N^3-78N^2+2168N-21792)$ |
| $C^{22}_{5153}$ |  | $\frac{1}{48}(3N^3-294N^2+10296N-130752)$ |
| $C^{22}_{5154}$ |  | $\frac{1}{48}(3N^3-318N^2+12072N-167184)$ |
| $C^{18}_{5155}$ |  | $\frac{1}{48}(4N^3-360N^2+11528N-133104)$ |
| $C^{18}_{5156}$ |  | $\frac{1}{48}(12N^3-1224N^2+44856N-599568)$ |
| $C^{22}_{5157}$ |  | $\frac{1}{48}(N^3-102N^2+3656N-46608)$ |

# Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

## 5 Vertices

$C^{24}_{5158}$  $\frac{1}{8}(3N^2 - 158N + 2248)$

$C^{20}_{5159}$  $\frac{1}{8}(8N^2 - 492N + 8112)$

$C^{16}_{5160}$  $\frac{1}{8}(10N^2 - 578N + 9024)$

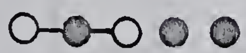
$C^{24}_{5161}$  $\frac{1}{8}(6N^2 - 414N + 7772)$

$C^{24}_{5162}$  $\frac{1}{8}(3N^2 - 220N + 4248)$

$C^{20}_{5163}$  $\frac{1}{8}(16N^2 - 1116N + 21512)$

$C^{16}_{5164}$  $\frac{1}{8}(20N^2 - 1386N + 26308)$

$C^{20}_{5165}$  $\frac{1}{8}(8N^2 - 528N + 9264)$

$C^{16}_{5166}$  $\frac{1}{8}(10N^2 - 688N + 12648)$

$C^{20}_{5167}$ 

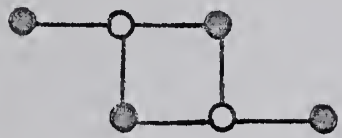
$C^{20}_{5168}$ 

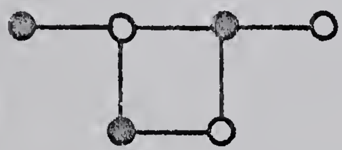
$C^{20}_{5169}$ 

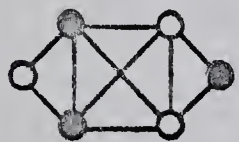
# Table Of LOW TEMPERATURE F.C.C. CONFIGURATIONS

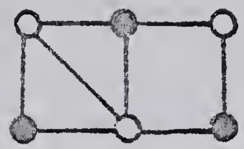
## 6 Vertices $\Delta E/2 = 10J, 12J$

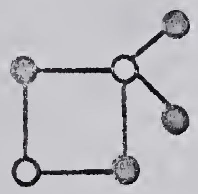
$C^{10}_{601}$  $\frac{5}{2}$

$C^{12}_{602}$  $\frac{29}{2}$

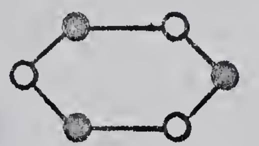
$C^{12}_{603}$  45

$C^{12}_{604}$  1

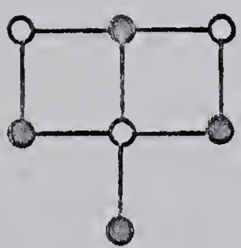
$C^{12}_{605}$  12

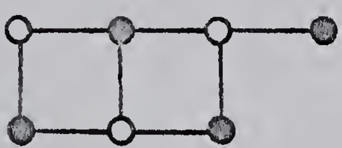
$C^{12}_{606}$  4

$C^{12}_{607}$  40

$C^{12}_{608}$  $\frac{11}{2}$

## 7 Vertices $\Delta E/2 = 12J$

$C^{12}_{701}$  $\frac{5}{2}$

$C^{12}_{702}$  15

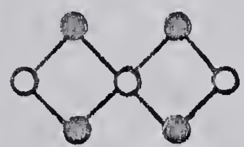
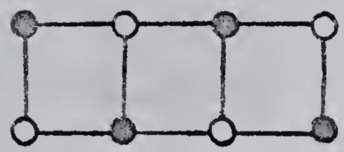
$C^{12}_{703}$  $\frac{15}{8}$

Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

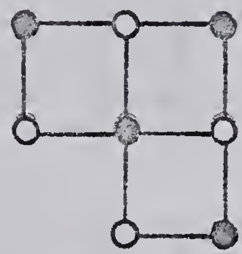
8 Vertices - $\Delta E/2 = 12J$

$C^{12}_{801}$



$\frac{9}{4}$

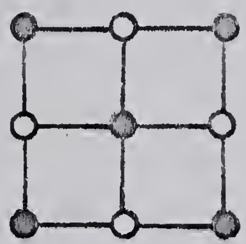
$C^{12}_{802}$



$\frac{19}{8}$

9 Vertices - $\Delta E/2 = 12J$

$C^{12}_{901}$



$\frac{9}{16}$

Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

6 Vertices $\Delta E/2 = 14J$

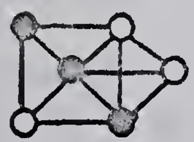
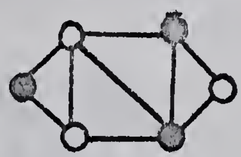
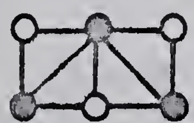
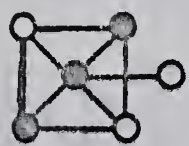
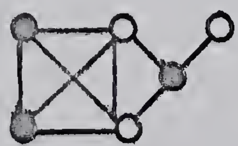
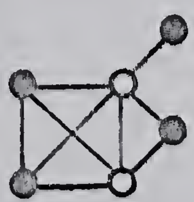
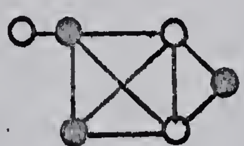
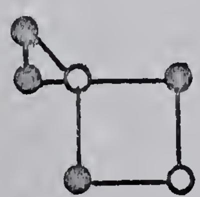
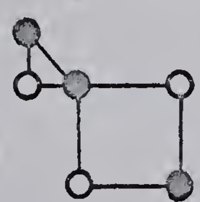
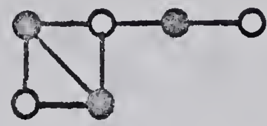
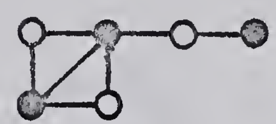
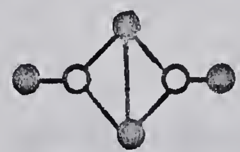
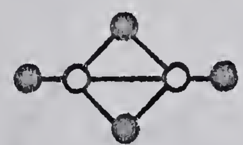
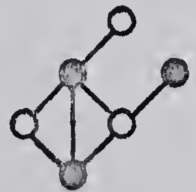
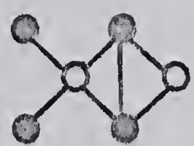
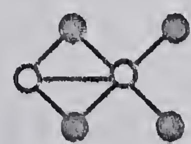
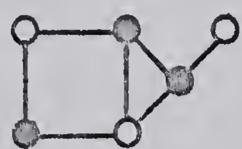
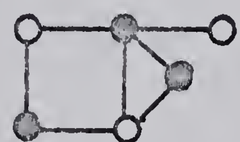
| | | |
|---|---|---|
| $C^{14}_{609}$ |  | 1 |
| $C^{14}_{610}$ |  | 6 |
| $C^{14}_{611}$ |  | 26 |
| $C^{14}_{612}$ |  | 7 |
| $C^{14}_{613}$ |  | 3 |
| $C^{14}_{614}$ |  | 16 |
| $C^{14}_{615}$ |  | 10 |
| $C^{14}_{616}$ |  | 16 |
| $C^{14}_{617}$ |  | 6 |
| $C^{14}_{618}$ |  | 6 |
| $C^{14}_{619}$ |  | 148 |
| $C^{14}_{620}$ |  | 72 |
| $C^{14}_{621}$ |  | 64 |
| $C^{14}_{622}$ |  | 17 |
| $C^{14}_{623}$ |  | 124 |
| $C^{14}_{624}$ |  | 20 |
| $C^{14}_{625}$ |  | 6 |
| $C^{14}_{626}$ |  | 18 |
| $C^{14}_{627}$ |  | 10 |
| $C^{14}_{628}$ |  | 14 |

Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

6 Vertices $\Delta E/2 = 14J$

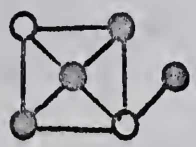
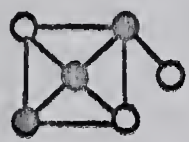
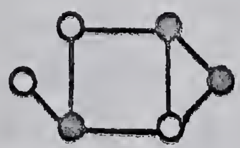
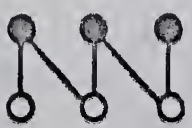
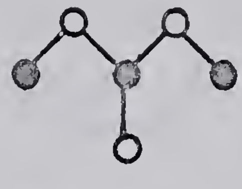
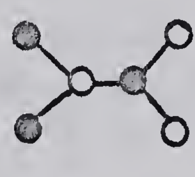
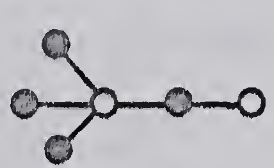
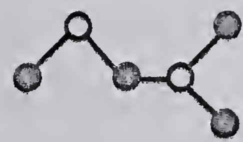
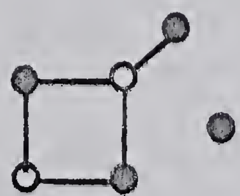
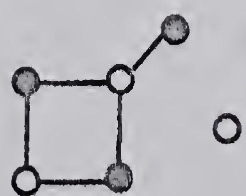
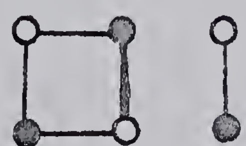
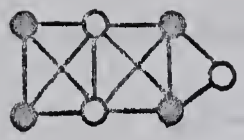
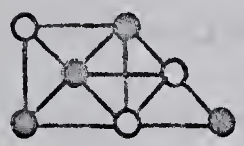
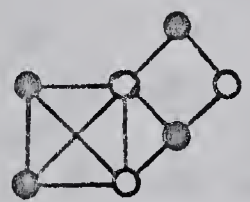
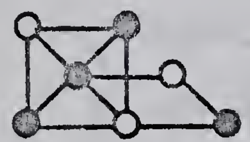
| | | |
|---|---|---|
| $C^{14}_{629}$ |  | 8 |
| $C^{14}_{630}$ |  | 11 |
| $C^{14}_{631}$ |  | 7 |
| $C^{14}_{632}$ |  | 22 |
| $C^{14}_{633}$ |  | 2082 |
| $C^{14}_{634}$ |  | $\frac{873}{2}$ |
| $C^{14}_{635}$ |  | $\frac{193}{2}$ |
| $C^{14}_{636}$ |  | 22 |
| $C^{14}_{637}$ |  | 492 |
| $C^{14}_{638}$ |  | $\frac{1}{2}(9N-305)$ |
| $C^{14}_{639}$ |  | $\frac{1}{2}(9N-310)$ |
| $C^{14}_{640}$ |  | $6N-253$ |

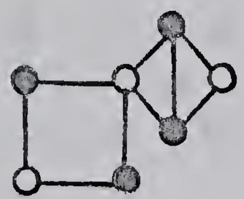
Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS
7 Vertices $\Delta E/2 = 14J$

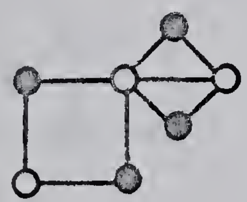
$C^{14}_{704}$  4

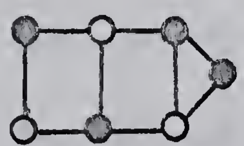
$C^{14}_{705}$  12

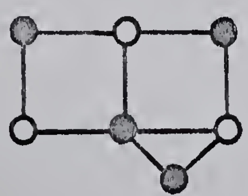
$C^{14}_{706}$  12

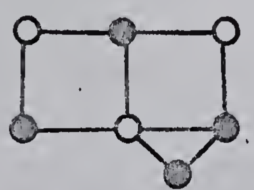
$C^{14}_{707}$  2

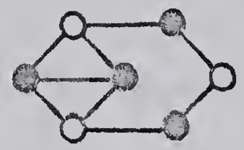
$C^{14}_{708}$  12

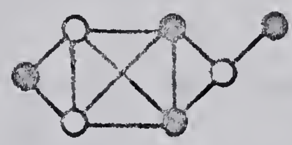
$C^{14}_{709}$  3

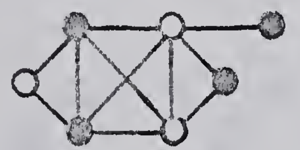
$C^{14}_{710}$  5

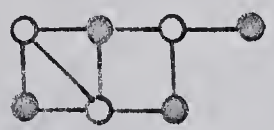
$C^{14}_{711}$  5

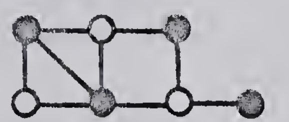
$C^{14}_{712}$  5

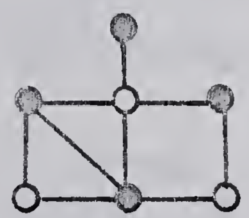
$C^{14}_{713}$  $\frac{3}{2}$

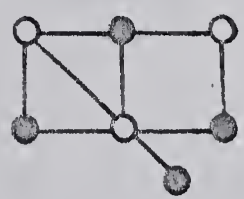
$C^{14}_{714}$  32

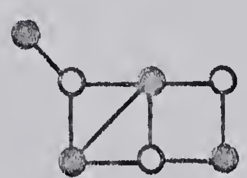
$C^{14}_{715}$  20

$C^{14}_{716}$  44

$C^{14}_{717}$  28

$C^{14}_{718}$  16

$C^{14}_{719}$  10

$C^{14}_{720}$  48

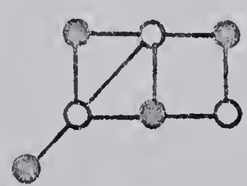
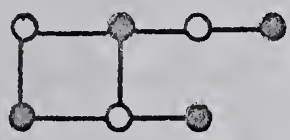
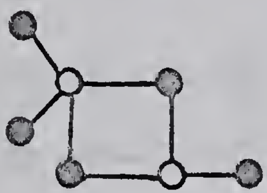
$C^{14}_{721}$  29

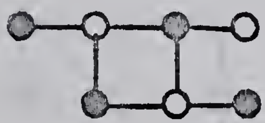
Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

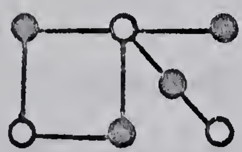
7 Vertices $\Delta E/2 = 14J$

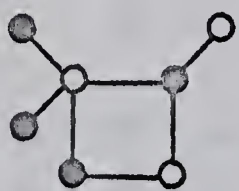
$C^{14}_{722}$  130

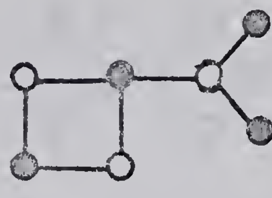
$C^{14}_{723}$  200

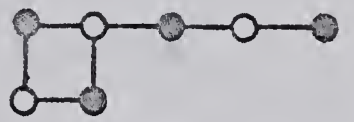
$C^{14}_{724}$  13

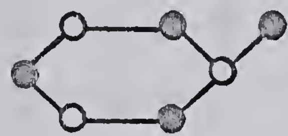
$C^{14}_{725}$  60

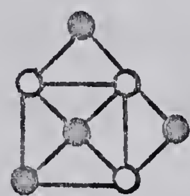
$C^{14}_{726}$  $\frac{55}{2}$

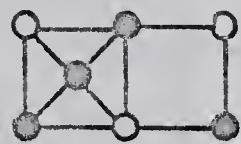
$C^{14}_{727}$  17

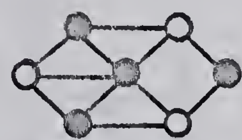
$C^{14}_{728}$  39

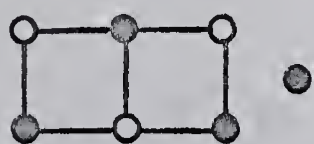
$C^{14}_{729}$  $\frac{381}{2}$

$C^{14}_{730}$  42

$C^{14}_{731}$  1

$C^{14}_{732}$  5

$C^{14}_{733}$  $\frac{7}{2}$

$C^{14}_{734}$  $\frac{5}{4}N - 50$

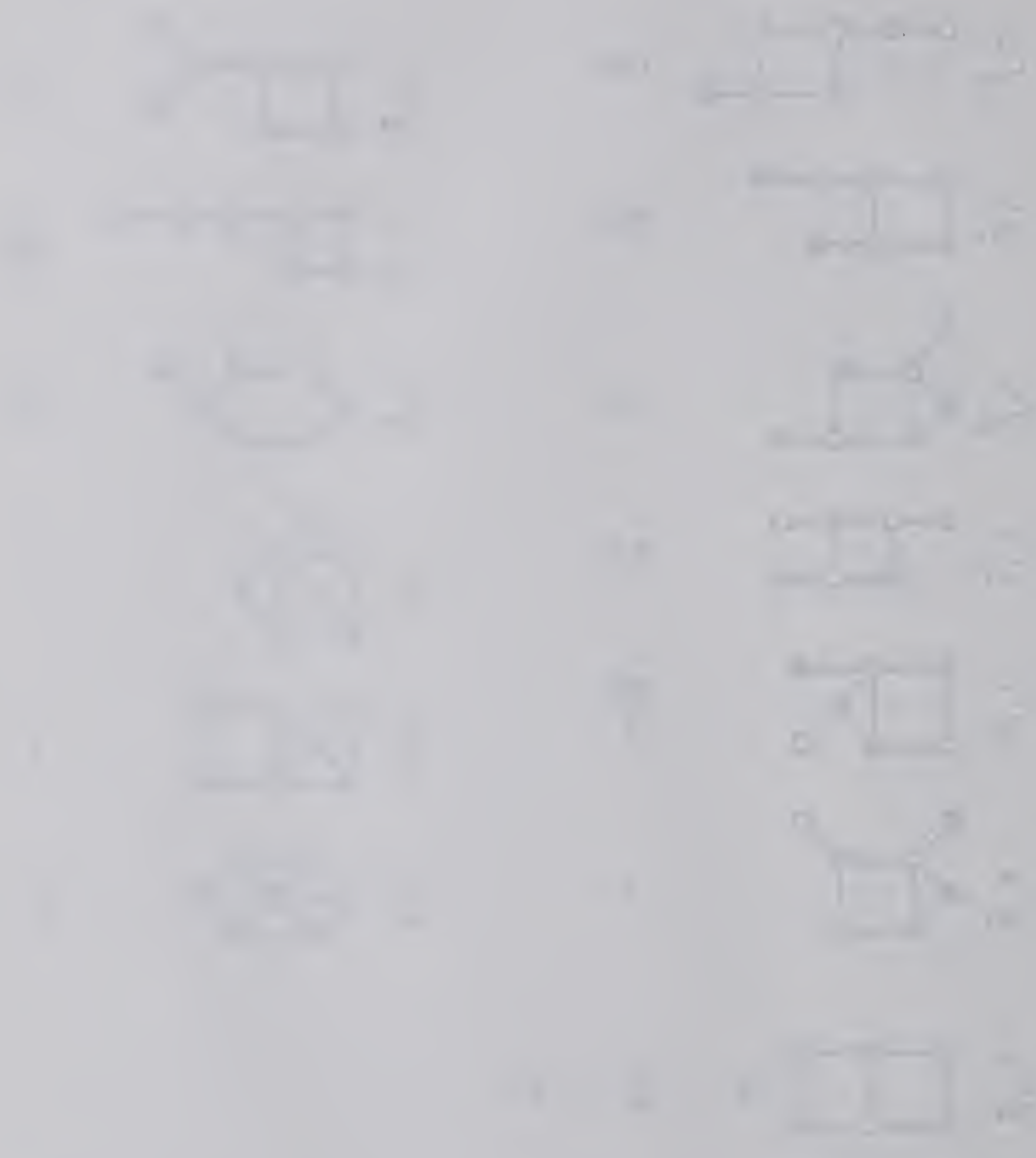
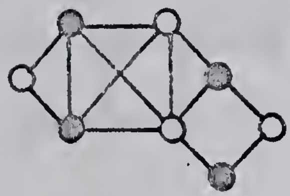
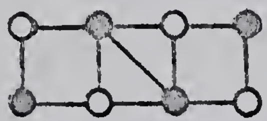


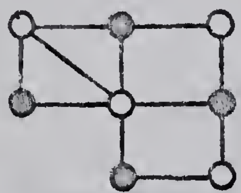
Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

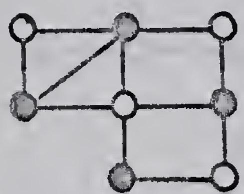
8 Vertices $\Delta E/2 = 14J$

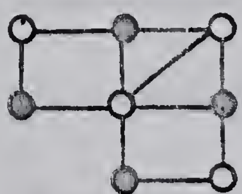
$C^{14}_{803}$  24

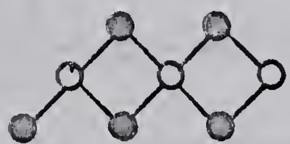
$C^{14}_{804}$  10

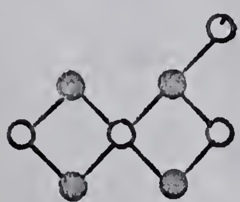
$C^{14}_{805}$  $\frac{5}{2}$

$C^{14}_{806}$  5

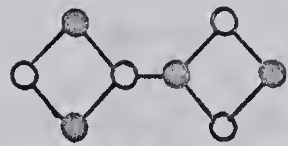
$C^{14}_{807}$  10

$C^{14}_{809}$  $\frac{5}{2}$

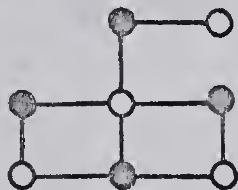
$C^{14}_{810}$  $\frac{25}{2}$

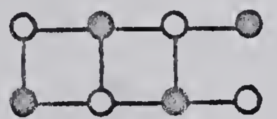
$C^{14}_{811}$  $\frac{81}{4}$

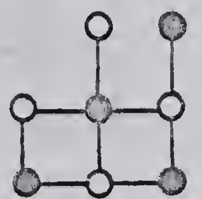
$C^{14}_{812}$  18

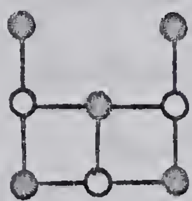
$C^{14}_{813}$  $\frac{33}{2}$

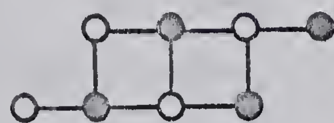
$C^{14}_{814}$  $\frac{263}{4}$

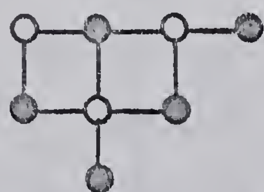
$C^{14}_{815}$  $\frac{31}{4}$

$C^{14}_{816}$  75

$C^{14}_{817}$  $\frac{41}{4}$

$C^{14}_{818}$  21

$C^{14}_{819}$  48

$C^{14}_{820}$  15

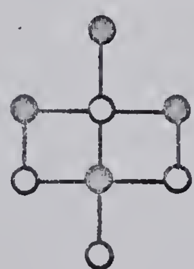
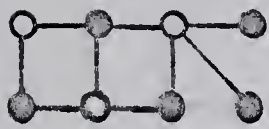
$C^{14}_{821}$  $\frac{5}{2}$

Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

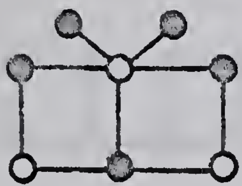
8 Vertices $\Delta E/2 = 14J$

$C^{14}_{822}$



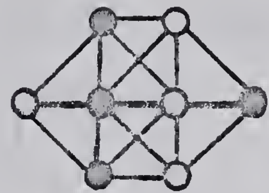
$8$

$C^{14}_{823}$



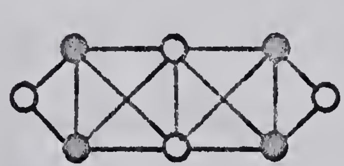
$0$

$C^{14}_{824}$



$\frac{7}{2}$

$C^{14}_{825}$



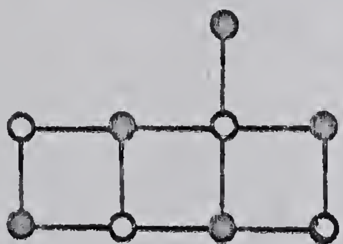
$4$

9 Vertices $\Delta E/2 = 14J$

$C^{14}_{902}$

$\frac{25}{4}$

$C^{14}_{903}$

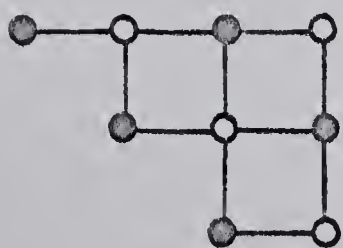


$\frac{27}{2}$

$C^{14}_{904}$

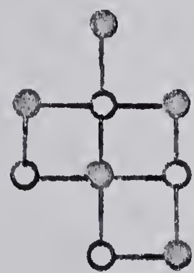
$\frac{9}{2}$

$C^{14}_{905}$



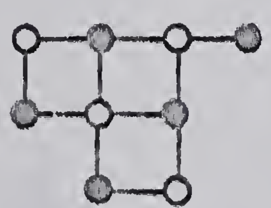
$\frac{29}{2}$

$C^{14}_{906}$



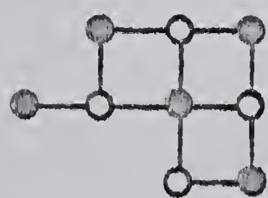
$\frac{19}{4}$

$C^{14}_{907}$



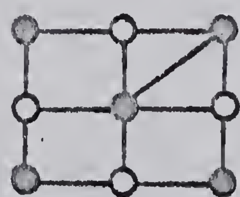
$7$

$C^{14}_{908}$



$\frac{19}{2}$

$C^{14}_{909}$

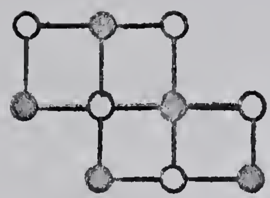


$\frac{5}{2}$

## Table of LOW TEMPERATURE F.C.C. CONFIGURATIONS

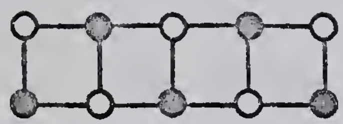
### 10 Vertices $\Delta E/2 = 14J$

$C^{14}_{1001}$



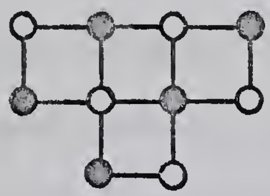
$\frac{37}{8}$

$C^{14}_{1002}$



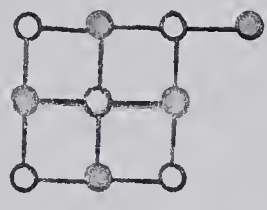
$\frac{17}{8}$

$C^{14}_{1003}$



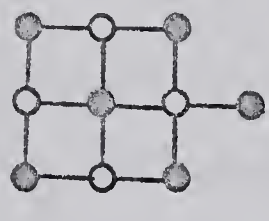
$\frac{9}{2}$

$C^{14}_{1004}$



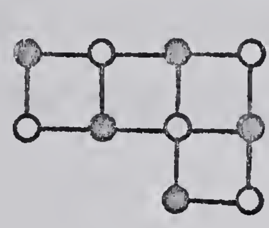
$\frac{27}{2}$

$C^{14}_{1005}$



$\frac{9}{4}$

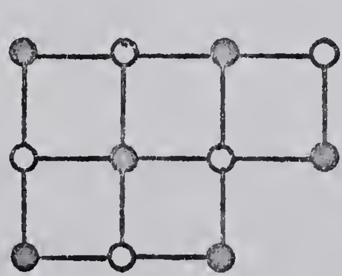
$C^{14}_{1006}$



$\frac{35}{8}$

### 11 Vertices $\Delta E/2 = 14J$

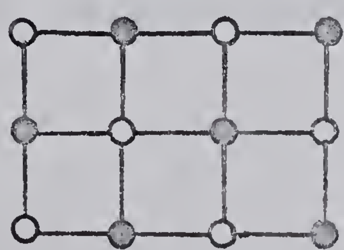
$C^{14}_{1101}$



$\frac{35}{8}$

### 12 Vertices $\Delta E/2 = 14J$

$C^{14}_{1201}$



$\frac{17}{8}$

# APPENDIX B.

## Counting Theorem

To prove equation (32) we proceed as follows: Consider a graph $X_1\ X_2$ -- $X_i$ -- $X_M$ of M components and the number of ways that this graph can be placed on the lattice so that two vertices of the tetrahedron are black and two are white is

$$[X_i]_M = \{X_i\}_M - \{\overset{\circ}{X_i}\}_M - \{\overset{\bullet}{X_i}\}_M - \{\overset{\circ}{X_i}\overset{\circ}{X_j}\}_M - \{\overset{\bullet}{X_i}\overset{\bullet}{X_j}\}_M$$

$$- \{\overset{\circ}{X_i}\overset{\bullet}{X_j}\overset{\bullet}{X_k}\}_M - \{\overset{\bullet}{X_i}\overset{\circ}{X_j}\overset{\circ}{X_k}\}_M - \{\overset{\circ}{X_i}\overset{\circ}{X_j}\overset{\bullet}{X_k}\overset{\bullet}{X_\ell}\}_M \tag{B.1}$$

Now first consider $\{\overset{\circ}{X_i}\}_M$

The number of ways $X_i$ can be rooted to a white vertex of the tetrahedron is

$$\frac{2\ \{X_i\}\ \ W_i}{\{o\}} = \frac{4}{N}\ \{X_i\}\ \ W_i$$

The factor 2 occurs because there are 2 white vertices on the tetrahedron; $W_i$ because each of the $W_i$ white vertices

of $X_i$ could be the root and there are $\dfrac{\{X_i\}}{\{o\}}$ ways of orienting $X_i$ on the root. However, we are not rooting $\{X_i\}$ but $\{X_i\}_M$. The number of ways this can be done is

$$\frac{4}{N}\{X_i\}\ W_i\ \frac{\{X_i\}_M}{\{X_i\}} = \frac{4}{N}\ W_i\ \{X_i\}_M$$

But in doing this some other of the M components are going to be rooted to the other three vertices. We must subtract these. So we get

$$\{\overset{\circ}{X}_i\}_M = \frac{4}{N} W_i \{X_i\}_M - \sum_{i,j} \{\overset{\circ}{X}_i\overset{\bullet}{X}_j\}_M - 2\sum_{i,j} \{\overset{\circ}{X}_i\overset{\circ}{X}_j\}_M$$

$$- 2\sum_{i,j,k} \{\overset{\circ}{X}_i\overset{\circ}{X}_j\overset{\bullet}{X}_k\}_M - \{\overset{\circ}{X}_i\overset{\bullet}{X}_j\overset{\bullet}{X}_k\}_M - 2\sum_{i,j,k,\ell} \{\overset{\circ}{X}_i\overset{\circ}{X}_j\overset{\bullet}{X}_k\overset{\bullet}{X}_\ell\}_M \qquad \text{(B.2)}$$

Similarly

$$\{\overset{\bullet}{X}_i\}_M = \frac{4}{N} B_i \{X_i\}_M - \sum_{i,j} \{\overset{\bullet}{X}_i\overset{\circ}{X}_j\}_M - 2\sum_{i,j} \{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\}_M$$

$$- 2\sum_{i,j,k} \{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\overset{\circ}{X}_k\}_M - \sum_{i,j,k}\{\overset{\bullet}{X}_i\overset{\circ}{X}_j\overset{\circ}{X}_k\}_M - 2\sum_{i,j,k,\ell}\{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\overset{\circ}{X}_k\overset{\circ}{X}_\ell\}_M \qquad \text{(B.3)}$$

Substitute B.2 and B.3 into B.1

$$
\begin{aligned}
[X_i]_M = {} & \{X_i\}_M - \frac{4}{N}\{X_i\}_M (B_i+W_i) + 2\sum_{i,j}\{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\}_M \\
& + 2\sum_{i,j}\{\overset{\circ}{X}_i\overset{\circ}{X}_j\}_M + 2\sum_{i,j}\{\overset{\circ}{X}_i\overset{\bullet}{X}_j\}_M + 3\sum_{i,j,k}\{\overset{\circ}{X}_i\overset{\circ}{X}_j\overset{\bullet}{X}_k\}_M \\
& + 3\sum_{i,j,k}\{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\overset{\circ}{X}_k\}_M + 4\sum_{i,j,k,\ell}\{\overset{\circ}{X}_i\overset{\circ}{X}_j\overset{\bullet}{X}_k\overset{\bullet}{X}_\ell\}_M
\end{aligned}
\tag{B.4}
$$

Now consider $\{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\}_M$

Clearly

$$\{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\} = \frac{\{X_i\}\{X_j\}}{\{\bullet\}^2}\, 2\, B_iB_j = \frac{8}{N^2}\{X_i\}\{X_j\}\, B_iB_j$$

$$\therefore \quad \{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\}_M = \frac{8}{N^2}\{X_i\}\{X_j\}\, B_iB_j\, \frac{\{X_i\}_M}{\{X_iX_j\}}$$

$$-\sum_{i,j,k}\{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\overset{\circ}{X}_k\}_M - \sum_{i,j,k,\ell}\{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\overset{\circ}{X}_k\overset{\circ}{X}_\ell\}_M \tag{B.5}$$

since $\dfrac{\{X_i\}_M}{\{X_iX_j\}}$ is the number of ways of putting the M-2 configurations on the lattice after fixing $X_i$ and $X_j$.

similarly

$$\{\overset{\circ}{X}_i\overset{\circ}{X}_j\}_M = \frac{8}{N^2}\{X_i\}\{X_j\}\, W_iW_j\, \frac{\{X_i\}_M}{\{X_iX_j\}}$$

$$- \sum_{i,j,k}\{\overset{\circ}{X}_i\overset{\circ}{X}_j\overset{\bullet}{X}_k\}_M \quad - \sum_{i,j,k,\ell}\{\overset{\circ}{X}_i\overset{\circ}{X}_j\overset{\bullet}{X}_k\overset{\bullet}{X}_\ell\}_M \qquad (B.6)$$

and

$$\{\overset{\circ}{X}_i\overset{\bullet}{X}_j\}_M = \frac{16}{N^2}\{X_i\}\{X_j\}\, W_iB_j\, \frac{\{X_i\}_M}{\{X_iX_j\}}$$

$$- 2\sum_{i,j,k}\{\overset{\circ}{X}_i\overset{\bullet}{X}_j\overset{\circ}{X}_k\}_M \quad - 2\sum_{i,j,k}\{\overset{\circ}{X}_i\overset{\bullet}{X}_j\overset{\bullet}{X}_k\}_M$$

$$- 4\sum_{i,j,k,\ell}\{\overset{\circ}{X}_i\overset{\bullet}{X}_j\overset{\circ}{X}_k\overset{\bullet}{X}_\ell\}_M \qquad (B.7)$$

Now substituting equations (B.5), (B.6) and (B.7) into (B.4)

$$[X_i]_M = \{X_i\}_M - \frac{4}{N}\{X_i\}_M\,(B_i + W_i)$$

$$+ \frac{8}{N^2}\{X_i\}_M \sum_{i,j}\frac{\{X_i\}\{X_j\}}{\{X_i\ X_j\}}\,(W_iW_j + B_iB_j + 2\ W_iB_j)$$

$$- 3\sum_{i,j,k}\{\overset{\circ}{X}_i\overset{\circ}{X}_j\overset{\bullet}{X}_k\}_M \quad - 3\sum_{i,j,k}\{\overset{\bullet}{X}_i\overset{\bullet}{X}_j\overset{\circ}{X}_k\}_M \quad - 6\sum_{i,j,k}\{\overset{\circ}{X}\ \overset{\circ}{X}\ \overset{\bullet}{X}\ \overset{\bullet}{X}\}_M$$

(B.8)

Now continuing in the same fashion.

$$\sum_{i,j,k} \{\overset{\circ}{X}_i \overset{\circ}{X}_j \overset{\bullet}{X}_k\}_M = \frac{2x2x8}{N^3} \quad W_i W_j B_k \frac{\{X_i\}\{X_j\}\{X_k\}\{X_i\}_M}{\{X_i X_j X_k\}} \tag{B.9}$$

$$- 2 \sum_{i,j,k,\ell} \{\overset{\circ}{X}_i \overset{\circ}{X}_j \overset{\bullet}{X}_k \overset{\bullet}{X}_\ell\}_M$$

$$\sum_{i,j,k} \{\overset{\circ}{X}_i \overset{\bullet}{X}_j \overset{\bullet}{X}_k\} = \frac{32}{N^3} W_i W_j W_k \quad \frac{\{X_i\}\{X_j\}\{X_k\}\{X_i\}_M}{\{X_i X_j X_k\}}$$

$$- 2 \sum_{i,j,k,\ell} \{\overset{\circ}{X}_i \overset{\circ}{X}_j \overset{\bullet}{X}_k \overset{\bullet}{X}_\ell\}_M$$

Substituting (B.9) and (B.10) into (B.8)

$$[X_i]_M = \{X_i\}_M - \frac{4}{N} \quad \{X_i\}_M \ (B_i + W_i)$$

$$+ \frac{8}{N^2} \quad \{X_i\}_M \sum_{i,j} \frac{\{X_i\}\{X_j\}}{\{X_i X_j\}} \quad (W_i W_j + B_i B_j + 2W_i B_j)$$

$$- \frac{96}{N^3} \quad \{X_i\}_M \sum_{i,j,k} \frac{\{X_i\}\{X_j\}\{X_k\}}{\{X_i X_j X_k\}} \quad (W_i W_j B_k + B_i W_j W_k)$$

$$+ 6 \sum_{i,j,k,\ell} \{\overset{\circ}{X}_i \overset{\circ}{X}_j \overset{\bullet}{X}_k \overset{\bullet}{X}_\ell\}_M$$

The last term in (B.11) is

$$\sum_{i,j,k,\ell} \{\overset{\circ}{X}_i \overset{\circ}{X}_j \overset{\bullet}{X}_k \overset{\bullet}{X}_\ell\}_M = \frac{2x2x16}{N^4} \quad \frac{\{X_i\}\{X_j\}\{X_k\}\{X_\ell\}}{\{X_i X_j X_k X_\ell\}} \{X_i\}_M$$

$$x\ W_i W_j B_k B \tag{B.12}$$

Finally we arrive at the result

$$[X_i]_M = \{X_i\}_M - \frac{4}{N} n_i \{X_i\}_M$$

$$+ \frac{8}{N^2} \{X_i\}_M \sum_{i,j} \frac{\{X_i\}\{X_j\}}{\{X_i X_j\}} (W_i W_j + B_i B_j + 2W_i B_j) \lambda_{ij}$$

$$- \frac{96}{N^3} \{X_i\}_M \sum_{i,j,k} \frac{\{X_i\}\{X_j\}\{X_k\}}{\{X_i X_j X_k\}} (W_i W_j B_k + B_i W_j W_k) \lambda_{ijk}$$

$$+ \frac{384}{N^4} \{X_i\}_M \sum_{i,j,k,\ell} \frac{\{X_i\}\{X_j\}\{X_k\}\{X_\ell\}}{\{X_i X_j X_k X_\ell\}} (W_i W_j B_k B\ ) \lambda_{ijk} \tag{B.13}$$

Where we have replaced $B_i + W_i$ by $n_i$

The $\lambda$ factors have been included to take care of identical components in the M component graph. It can be shown that

$$\lambda_{ij} = 1 \qquad X_i \not\equiv X_j$$

$$= \frac{1}{2!} \qquad X_i \equiv X_j$$

$$\lambda_{ijk} = 1 \qquad X_i \not\equiv X_j \not\equiv X_k$$

$$= \frac{3}{8} \qquad X_i \equiv X_j \quad X_i \not\equiv X_k$$

$$= \frac{1}{3!} \qquad X_i \equiv X_j \equiv X_k$$

$$\lambda_{ijk\ell} = 1 \qquad X_i \not\equiv X_j \not\equiv X_k \not\equiv X$$

$$= \frac{5}{8} \qquad \text{(2 of X's identical)}$$

$$= \frac{1}{4} \qquad \text{(3 X's identical)}$$

$$= \frac{1}{16} \qquad \text{(4 X's identical)}$$

For M = 1 only the first two terms in (B.13) are non zero and the calculation involved is trivial.

For M = 2 the third term becomes non zero but it can be simplified as follows:

$$[X_1X_2] = \{X_1X_2\} - \frac{4n}{N} \{X_1X_2\}$$

$$\frac{8}{N^2} \{X_1\}\{X_2\} [n_1n_2 + 3(W_1B_2 + B_1W_2)] \lambda_{12}$$

The corresponding counting theorem for the two spin correlation parameter $R_2$ is, of course, simpler in form and is

$$[X_i]_M = \{X_i\}_M - \frac{2}{N} n_i \{X_i\}_M + \frac{4}{N^2} \{X_i\}_M \sum_{i,j} \frac{\{X_i\}\{X_j\}}{\{X_iX_j\}} (W_iB_j + B_iW_j) \lambda_{ij} \qquad (B.15)$$

# APPENDIX C.

## Padé Approximants

### C.1 Long Range Order Approximants

$$\frac{d \log (R_2)}{d(y^2)} = - 8y^2 - 96y^4 - 840y^6 - 7140y^8 - 62585y^{10} - 571459y^{12}$$

$$[1,1] = \frac{- 8y^2 - 26y^4}{1 - 8.75y^2}$$

$$[2,2] = \frac{- 8y^2 - 100.1905y^4 - 258.619y^6}{1 + .5238095y^2 - 78.95833y^4}$$

$$[1,3] = \frac{- 8y^2 - 25.87672y^4 + 1.47936y^6 + 222.9444y^8}{1 - 8.76541y^2}$$

$$[3,1] = \frac{- 8y^2 + .16912y^4}{1 - 12.02114y^2 - 39.25368y^4 + 840.26386y^6}$$

$$[2,3] = \frac{- 8y^2 + 70.697625y^4 + 339.4893y^6 + 512.6643y^8}{1 - 20.8372y^2 + 102.61027y^4}$$

$$[3,2] = \frac{- 8y^2 + 54.8391y^4 + 140.8803y^6}{1 - 18.85489y^2 + 103.64863y^4 - 156.52025y^6}$$

$$[4,1] = -\left[\frac{8y^2 + 6717.6471y^4}{1 - 851.70588y^2 + 10115.4766y^4 - 32489.0294y^6 + 84388.31618y^8}\right]$$

$$[1,4] = -\left[\frac{8y^2 - 22.95259y^4 + 36.568890y^6 + 529.977790y^8 + 2609.8112167y^{10}}{1 - 9.1309259y^2}\right]$$

$$[5,0] = -\left[\frac{8y^2}{1 - 12y^2 + 3900y^4 - 100.5y^6 - 2.125y^8 - 1784.375y^{10}}\right]$$

C.2 Specific Heat Approximants

$$\frac{d}{dy} \ln \left[\frac{1}{(\log y)^2 \; 16y^4} \quad \frac{Cv}{k}\right]$$

$$= 9 + 120y^2 + 393.375y^4 + 3792y^6 + 42224.47y^8$$

$$[4,0] = \frac{9}{1 - 13.3333y^2 + 139.06944y^4 - 1626.14815y^6 + 16748.19334y^8}$$

$$[3,1] = \frac{9 + 92.69373y^2}{1 - 3.0340299y^2 - 3.25460245y^4 - 245.3262475y^6}$$

$$[2,2] = \frac{9 + 12.8480136y^2 - 968.460579y^4}{1 - 11.9057766y^2 + 7.4286240y^4}$$

$$[1,3] = \frac{9 + 19.7836946y^2 - 942.8424y^4 - 588.28768y^6}{1 - 11.135145y^2}$$

C.3 Zero Field Susceptibility Approximants

$$\frac{d}{dy}\left(\frac{kT}{(2m^2)}\right) \chi_o = 2(2y^3 + 40y^7 + 90y^9 + 1691.25y^{11} + 10848.25y^{13} + \text{-------})$$

$$[1,4] = y^3\left[\frac{2 - 12.828677y^2 + 40.0000y^4 - 166.5735403y^6 + 1113.95953y^8}{1 - 6.4143385y^2}\right]$$

$$[2,3] = y^3\left[\frac{2 - 9.4615449y^2 - 23.274024y^4 - 99.230898y^6}{1 - 4.7307725y^2 - 31.63701196y^4}\right]$$

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